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

Annual Progress Report, May 1982 to May 1983

June 1983

**Prepared for the U.S. Nuclear Regulatory Commission
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PACIFIC NORTHWEST LABORATORY
operated by
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under Contract DE-AC06-76RLO 1830

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

ANNUAL PROGRESS REPORT
MAY 1982 TO MAY 1983

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Pacific Northwest Laboratory
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EXECUTIVE SUMMARY

This report documents progress for four major research projects which include 10 discrete research tasks being conducted for the U.S. Nuclear Regulatory Commission (NRC), Office of Research, Waste Management Branch. The primary purpose of these tasks is to provide information to help the NRC license uranium recovery facilities. A truncated title of each task and a brief highlight summary are provided here.

● Long-Term Stabilization

The baseline data studies on long-term stabilization techniques were completed and reports issued in four categories: 1) rock riprap design methods, 2) wind erosion, 3) overland erosion, and 4) survivability of ancient earthen mounds. The theory and currently accepted design procedures for riprap design methods were reviewed; the physical processes of wind and water erosion, and the models used to estimate erosion rates and soil loss were reviewed; and ancient man-made earthen mounds were evaluated as approximate analogs of uranium mill tailings impoundments.

Using the baseline studies, an interim handbook for riprap design was compiled and forwarded for review and comment by NRC staff. The comments were used to revise the subtask work for FY-83.

Field trips were made to Mt. Sopris, Colorado, to gather rock samples with known weathering ages for rock durability studies, and to several uranium mill tailings sites to select two disposal sites that will be analyzed in detail during the technical and economic feasibility study. The impoundments at Slickrock Mill (Colorado) and Shiprock (New Mexico) were tentatively selected for analysis.

The use of ultra-sound for predicting rock durability showed little promise in laboratory studies of a suite of rock samples with the same lithologic source for different weathering ages. Future studies will use semi-quantitative, geomorphologic field techniques coupled with more conventional rock-durability tests.

Five papers representing work in this task were presented at the Fifth Symposium on Uranium Mill Tailings Management in Fort Collins, Colorado, on December 9, 1982. The presentations discussed the results of the baseline data studies.

● Interim Stabilization of Mill Tailings Piles

The interim stabilization task activities were directed toward identifying and evaluating the materials that are commercially available to control fugitive dust from uranium tailings. Both laboratory and field studies were conducted. Laboratory studies consisted primarily of wind erosion tests conducted in a wind tunnel under a variety of accelerated weathering conditions.

More than 40 chemical stabilizers were tested. Field studies consisted of field testing the more promising stabilization techniques and monitoring the effectiveness of the stabilizers. During August 1982, 15 chemical stabilizers were applied to a test plot at a uranium mill site in Gas Hills, Wyoming. Periodic visits to the test site documented the weathering of the various stabilizers primarily by photographing the test site. A portable weather station is accumulating meteorological data to document the weathering conditions that the stabilizers experience. Additional field testing is planned for August 1983, and will include both chemical stabilizers and windscreen systems.

- Tailings Dewatering Techniques

The classical consolidation theory that is available in the variably-saturated flow and consolidation model, TRUST, has been determined inadequate for the analysis of hydraulically filled tailings pits. A more recent theory developed by Fredlund and Morgenstern is a better description. This new theory, which is based on a two stress-state description of the soil void ratio in the unsaturated zone, is incorporated into the model algorithm (now called TRUST-II). The effective-stress representation of the stress state of the system will still be used in the saturated zone.

The experimental apparatus for determining the soil properties that are necessary to perform simulations of fluid flow and consolidation in uranium mill tailings has been obtained and the equipment is currently being set-up and modified to perform the necessary laboratory measurements. In addition, methods for preparing input to TRUST-II and for analyzing the results have been developed. This package of routines contains several programs for developing numerical grids, preparing soil characteristic data, and for analyzing the transport of contaminants through the flow field generated by TRUST-II.

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

A literature review was conducted to evaluate the amelioration techniques currently used in various mining industries to immobilize toxic materials. Neutralization, fixation, and specific constituent removal were identified as possible methods for treating uranium mill tailings solutions. Neutralization processes appear to be the best suited for treating uranium mill tailings. In general, fixation processes are not as applicable because they are expensive and increase the weight and volume of waste. Specific removal processes are currently practiced only for ^{226}Ra . Many of the specific removal techniques are either 1) still being researched, 2) more costly than other techniques, 3) less efficient, or 4) unproven for use in acidic tailings liquor.

Laboratory experiments were conducted to evaluate selected neutralizing agents for use on uranium tailings solutions. Laboratory results indicate that either calcium hydroxide or its dehydrated form (lime) appears to be the most effective treatment option.

A plan was submitted for a field demonstration to evaluate the effectiveness, costs, and benefits of neutralization of acid uranium mill tailings in order to reduce the potential leaching of toxic trace elements, radionuclides, and macro ions from a tailings impoundment.

- Disposal Techniques Allowing Contact with Ground Water

The potential environmental concerns that might result from disposal of uranium tailings below the water table are being evaluated in this task.

Results of the evaluation show that the leachate from uranium mill tailings in disposal pits is best controlled by a combination of a clay bottom liner, clay stub-sidewall liners, and tailings drains that collect effluent for subsequent pumping from the pit. As much as 80 percent of the total leachate volume is intercepted by this combination of control features that otherwise would enter ground water from disposal pits above the water table. Model simulations of pits below the water table equipped with the same combination of controls showed essentially no leachate loss. Pumping of the effluent, however, involves continuing energy costs and requires an alternative method of disposal of any leachate that cannot be reused as mill process-makeup water.

When no drains or effluent pumping are involved, the clay bottom liners are less useful. The liners reduce the leachate outflow rates, but the leachate continues to flow out over a much longer period of time, and this results in essentially the same total outflow volume of leachate. This pattern is similar for disposal both above and below the water table, although more leachate enters the ground water from disposal pits below the water table.

Current efforts are directed to assess potential environmental hazards associated with disposal of tailings below the water table in abandoned deep mine stopes. This work will be completed by the end of the first quarter of FY-84.

- Effluent and Environmental Monitoring Methods and Equipment and Instrument Testing

To assess the accuracy of short-term measuring devices for radon and radon-daughters, the following short-term measurements were compared to long-term measurements of indoor radon-daughter concentrations in buildings in Edgemont, South Dakota: single, five-minute, air filter measurements; single 100-hr Radon Progeny Integrated Sampling Unit (RPISU) measurements; two 100-hr RPISU measurements taken six months apart; and two-month, six-month, and twelve-month integrated Terradex Track Etch® measurements. Results of these comparisons indicate how accurately different measuring techniques predict Radon Progeny Integrated Sampling Unit (RPISU) average annual radon-daughter concentrations. Further analyses of data from Edgemont showed no appreciable linear correlation between indoor radon-daughter concentrations and 1) wind

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speed, 2) barometric pressure, 3) barometric pressure changes, 4) outdoor temperature, and 5) outdoor radon or radon-daughter concentrations. Although a small linear correlation was found between calculated turnover times for radon daughters and wind speed for the same, previous, and second previous hours during which the measurements of radon-daughter concentrations used to calculate the turnover time were taken, building-to-building variations overshadowed variations in concentrations caused by wind speed. Results indicate that because building-to-building variations are so large, it will be necessary to take a large number of measurements over time in a limited number of buildings in order to accurately determine the effects of ambient conditions on indoor radon-daughter concentrations. Other work evaluated the accuracy of various long-term measuring devices for radon and radon-daughter.

Results of the other subtasks in Task 6 address the techniques and procedures used to measure concentrations of certain effluents and verify the decontamination of sites and structures, and are listed below:

- Recommendations have been extracted from available EPA and Argonne National Laboratory literature for monitoring of yellowcake emissions. The procedures that are currently in use to monitor yellowcake emissions at various uranium mills have been determined.
- Existing methods of wet chemical analysis are adequately sensitive to meet the detection limits set in NRC Regulatory Guide 4.14, but are costly and time-consuming. Alternatives have been investigated.
- Many of the tools and methods that are used in borehole logging by the uranium exploration industry can be applied to remedial action projects to measure uranium and its progeny.
- Existing analytical methods for detecting thorium radium, selenium, arsenic, antimony, zinc, nickel chromium, and copper in surface and ground water, satisfactorily meet the monitoring needs anticipated at uranium recovery sites. Sampling protocol is being statistically evaluated.

- Attenuation of Radon Emissions

Additional work was completed this year to better-define methods for determining the thickness of cover soils needed to reduce radon flux from uranium-mill tailings to prescribed limits. Diffusion coefficients for radon in 34 candidate cover soils ranged from 0.001 to 0.06 cm²/s, depending on the type of soil and its expected moisture content at the tailings site. Radon diffusion coefficients evaluated in the laboratory with 0.1-m depths of soil were demonstrated to be statistically indistinguishable from those evaluated with several-fold larger depths of the same soils used to cover uranium-mill tailings. An alternative laboratory method for measuring radon diffusion coefficients in soil was developed, and it was established that this method validated the coefficients obtained by other methods. A mathematical model was also developed that predicted the values of experimentally measured radon diffusion coefficients in soil to within a factor of 6, using only the moisture content and pore-size distribution of the soil as parameters. Research was

also largely completed on evaluating the advective transport of radon through soil covers and on examining the influence of cover defects on radon flux from the cover surface.

This project is scheduled for completion in FY-83. During the remaining time, a handbook for determining radon attenuation through cover soils will be revised based on the results of research carried out since its preparation, a study will be completed to test the validity of methods recommended in the handbook, and a document will be prepared in the format of a Nuclear Regulatory Guide which specifies acceptable methods for covering uranium-mill tailings with soil to reduce radon flux to prescribed limits.

- Assessment of Leachate Movement

The laboratory analyses and predictive geochemical modeling in this study were conducted to assess long-term environmental impacts of leachate movement from uranium mill tailings. Results of experiments in the laboratory to determine changes in liner materials (materials lining the uranium impoundment to contain leachates) showed that liner failure (i.e., an increase in the permeability of the liner material) was not a problem when various acidic tailings solutions were leached through liner materials. Liner materials with a high clay content decreased in permeability over time. This is attributed to pore plugging resulting from precipitation of minerals and solids, and to soil particle dispersion. Initial work was also completed on the selection of analytical methods for determining uranium mill chemistry. Recommended analysis techniques and practical detection limits were determined for high-acid, high-salt tailings solutions because these solutions are difficult to analyze for metals.

Uranium mill tailings solutions spiked with radioactive tracers were percolated through columns packed with sediments and clay liner material and the resulting empirical data on trace contaminants suggest that:

- arsenic, chromium, lead, silver, thorium and vanadium are readily immobilized by the sediments and clay liner and are not released if acid conditions again prevail
- cadmium, iron, manganese, nickel, zinc, similar trace metals and uranium are immobilized by the sediments and clay liner as long as the solution pH is not driven below pH 4.5 by continual acid solution attack
- portions of selenium and molybdenum are very mobile and travel with the same velocity as the water
- laboratory results are capable of being extrapolated to actual field disposal conditions.

Geochemical modeling was begun using the mass transfer capabilities of the MINTEQ code. The code was used in a predictive mode to simulate the pH-front breakthrough and effluent compositions of uranium mill tailings percolating through the Morton Ranch clay liner in both permeability and vacuum

extractor columns. This work attempted to validate the chemical model postulated for a system involving interaction of geologic sediments with uranium mill tailings.

An initial conclusion drawn from these studies is that the laboratory experiments and geochemical modeling predictions were capable of simulating field observations. The same mineralogical changes and contaminant reductions observed in the laboratory studies were found at a drained evaporation pond (Lucky Mc Pathfinder in Wyoming) with a 10-year history of acid attack. The observed minerals were also predicted by geochemical (thermodynamic) modeling to form upon neutralization of the acidic tailings solutions by the contacted sediments. Thus, the laboratory studies, field observations, and theoretical model predictions complement each other.

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

The In-Situ Project began in October 1981. During the first six months, literature on aquifer restoration regulations, techniques, and practices was surveyed. Ground-water, lixiviant, and aquifer-sediment samples were collected from an operating mine in South Texas. Throughout the past year, these samples were used in laboratory experiments to determine the capacity of the natural aquifer system to restore the ground-water chemistry to its pre-mining state. Although the results of the experiments strictly apply only to the site sampled, the chemical processes identified as active in this aquifer should be representative of deep, confined aquifers in most places where uranium roll-front deposits occur. The experiments have shown that:

- Batch tests and, particularly, flow-through column experiments adequately simulate the geochemical environment at depth in an ore-zone aquifer. These experiments can determine some of the important chemical reactions that will occur between lixiviant and aquifer sediment.
- The concentration of uranium in solution, and by inference the concentration of other redox-sensitive contaminants (e.g., As, Se, and Mo), will be lowered by adsorption and the formation of secondary minerals and amorphous solids.
- The concentration of major cations (Na, Ca, Mg, and K) in solution may be changed by ion exchange, but their concentration in solution will not be limited by the formation of a solid phase, nor will the total dissolved solids level of the leaching solution be appreciably changed by interactions with the sediments.
- The fate of major anions in solution varies by constituents. Chloride concentrations are not expected to be affected by chemical processes in the aquifer. Carbonate concentration appears to be limited by the formation of carbonate minerals, especially calcite. The concentration of dissolved sulfate was not affected by solution/sediment interactions occurring in our experiments.

- Based on the changes in solution composition noted in the flow-through tests and on the typical concentration of ground water surrounding the uranium ore zone, the chloride and sulfate concentrations and total dissolved solids level of water in monitoring wells would be good indicators of excursions of the lixiviant from the leach field.

Additional experiments on natural restoration were begun to test its ability to retard the migration of As, Se, and Mo in the ore-zone aquifer. Also, induced restoration techniques are being evaluated and include both injection of ground water treated with chemical reductants and recirculation with surface treatment techniques such as reverse osmosis and electrodialysis. The methods will be tested as part of a field demonstration if it is deemed necessary.

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INTRODUCTION

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

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

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

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

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

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

PNL's program (Figure 1) consists of four coordinated projects comprised of a program management task and nine research tasks that address the critical technical and safety issues for uranium recovery. Specifically, the projects endeavor to find and evaluate methods to:

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

This is the first annual progress report for the uranium recovery research program at PNL, and covers the research performed for the NRC at PNL in the period May 1982 through May 1983 (this report takes the place of the progress report for the second quarter of FY-83). The following sections report the progress in uranium recovery research at PNL during the year, and briefly summarize the uranium recovery research conducted this past year through the Department of Energy's Uranium Mill Tailings Remedial Action Program.

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

FIGURE 1. Uranium Recovery Research for NRC at Pacific Northwest Laboratory

TASK 1. LONG-TERM STABILIZATION

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FIN B-2370 (Task 1)

OBJECTIVE

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

ACCOMPLISHMENTS DURING PAST YEAR

We have reported our accomplishments below using the subtask breakdown in effect during the first and second quarters of FY-83. The baseline data studies and the development of an interim handbook, which began in FY-82, are not listed as subtasks.

Baseline Data Studies

1. Rock Riprap Design Methods

During this study, we reviewed the available methods for design of rock riprap covers and their development, and we provide a preliminary assessment of their applicability for long-term protection of uranium mill tailings impoundments against flood erosion. We also considered other items regarding the application of riprap (e.g., placement, causes of failures, rock gradation, and rock durability).

Rock riprap design methods have been developed around the concept of the initiation of motion of a single particle. Hydrodynamic lift and drag are the primary forces acting on a rock particle subjected to flowing water. The submerged weight of the particle resists these forces. When motion of an individual rock is about to occur, the lift and drag forces are just balanced by the resisting force (submerged weight) of the rock. This is the condition of incipient motion. Three basic approaches are used by engineers to explain and describe the beginning of particle motion: 1) critical velocity approach, 2) critical shear-stress approach, and 3) lift-force mechanism.

The critical velocity approach considers a force balance on a rock particle where the flow imposes both a lift and drag force that tends to dislodge and entrain the particle. This approach lacks a good definition of an appropriate velocity and its measurement.

The critical shear-stress approach considers a force balance on a water prism and the resulting shear force over the particle. This approach has been popular because a critical shear stress can be easily related to flow parameters.

The lift-force mechanism involves a pressure difference on the particle, as well as upward velocity components resulting from turbulence. The pressure difference is the result of the steep velocity gradient along the rock particle. Most of the work in this area has been an attempt to understand and describe the lift forces on particles, but has not provided any critical lift criteria. Another drawback is that drag forces are ignored.

Most riprap design methods were developed by federal and state agencies for application to particular public works projects. Typical projects requiring riprap protection are highway embankments, bridges, flood channels, canals, and stilling basins. The type of project to be protected and the experience of each agency greatly influence the method chosen.

We reviewed six riprap design methods in this study. The first method reviewed is one that was recently developed by Colorado State University for the Wyoming Highway Department. This method incorporates the concept of a design safety factor and provides a much more versatile approach than the other methods. A stone size is determined through sets of equations developed for four basic flow conditions. This method offers the most promise as a procedure to be used to design riprap for the protection of tailings impoundments. It is usually referred to as the Safety Factor Method.

Two other methods were reviewed in detail to illustrate typical approaches to riprap design. These are the design procedures recommended by the U.S. Army Corps of Engineers and the California Highway Department. Both procedures use graphical techniques developed from basic equations to solve for the required stone sizes. This step in the design procedure is relatively simple. The rest of the design procedure is guided by written specifications and relies heavily on engineering experience. Three other methods that were briefly reviewed for comparison are the U.S. Bureau of Reclamation and the U.S. Bureau of Public Roads design methods, and the recommendations of the American Society of Civil Engineers Sedimentation Manual.

The application of a riprap design method to the long-term protection of tailings impoundments depends on the impoundment location in the drainage basin. Tailings impoundments located in upland watershed areas experience flooding to a lesser extent. The riprap design methods presented in this report would normally be used to design riprap for flood protection and not for overland erosion. However, the Safety Factor Method can be used to assist in designing riprap on side slopes when no lateral flow is expected.

The requirement for long-term protection of decommissioned tailings impoundments introduces three problem areas: 1) the effect of climate changes on a design flood magnitude, 2) local scour and hydrodynamic forces on the structure during floods, and 3) rock durability over long time periods.

Flood calculations used in design may have to allow for shifts in the climatic regime. This is based on the fact that a much wetter climate than that of today existed only about 150 years ago and extended back in time for several hundred years. The probable maximum flood (PMF) method is recommended for the determination of a design flood but may need to be modified to accommodate climatic changes.

Nonuniform flow regions are caused by the irregular shape of a tailings impoundment opposing the passage of a flood wave. This could result in local scour which could severely damage or undermine the riprap protection. This type of problem is usually investigated through laboratory flume studies.

Rock durability is a measure of the resistance of the rock material to the process of mechanical weathering or physical disintegration, and chemical weathering or decomposition. Several tests are available to assist in selecting better-quality rock, but they do not provide an estimate of how long a rock will resist weathering or remain durable.

2. Wind Erosion and Soil Deposition Models

The mechanics of wind erosion and soil deposition, and several erosion models were reviewed in this study. We evaluated the applicability of the models in estimating the erosion of soil from a tailings cover.

The model that best describes the wind erosion from a tailings pile is the wind erosion equation developed by W. S. Chapil. Chapil's equation contains more factors describing the variables that influence wind erosion from a tailings pile than any other model reviewed. Charts and graphs for obtaining the various factors in the equation, and the development of a computerized program make the equation fairly easy to use.

The amount of soil lost from a tailings pile as a result of wind erosion over many years can be calculated using the wind erosion equation. For a tailings pile in western Colorado, an unprotected soil surface can erode at a rate of almost 2.5 cm/yr. However, for any particular year, the amount of soil lost can vary depending on the wind and precipitation for any particular location. If considered on a seasonal basis, the variations from average values would be greater; one-way variations from average values would be even greater. To improve the accuracy of the equation and to estimate the variability of the annual values predicted, the probability functions for some of the dynamic variables could be incorporated.

Many methods for decreasing the soil erosion from a tailings pile will increase soil deposition. The use of wind breaks or riprap are two such methods. Increasing the surface roughness of a tailings pile with riprap will cause wind-blown soil to fill in the spaces between the riprap. Soil deposited over long time periods will increase the surface cover, and such an increase

has both positive and negative implications. Changes in the distribution of soil moisture could enhance revegetation. However, some chemical effects from soil interacting with riprap material could be detrimental to stability of the rock.

3. Overland Erosion and Computational Methods

We reviewed the overland erosional processes resulting from rainfall-runoff, and the computational methods currently used to predict soil losses and sediment yields from overland flow. A summary of the study results follows:

Surface runoff and erosion involves three basic processes: 1) infiltration, 2) overland flow, and 3) throughflow. Infiltration is the vertical movement of water through the soil surface and into the deeper layers. The major variables influencing the infiltration capacity of a soil are soil texture, thickness, degree of compaction, and soil-moisture content.

Overland flow, which travels over the ground surface, can be divided into two different types. Horton overland flow is primarily caused by nearly impermeable surface soils that greatly reduce infiltration. Saturated overland flow occurs because of saturated soil conditions. Horton overland flow is more common in the semi-arid western states and saturated overland flow is predominant in the eastern U.S.

The highest annual sediment yields in the U.S. occur typically in the semi-arid regions. Although these regions only experience from 10 to 15 inches of precipitation, high erosion rates occur because of high-intensity, short-duration thunderstorms and a lack of vegetative cover. The more humid regions have a higher and more even distribution of rainfall and can support a thicker vegetation cover. The implications are that decommissioned uranium tailings impoundments located in semi-arid areas will experience the most severe erosion problems relative to other areas, and that Horton overland flow will be the primary cause of soil loss.

The computation methods for overland flow and erosion include both regression and physical-process simulation models. The most widely used regression model is the Universal Soil Loss Equation (USLE), which determines soil losses on an annual basis. The physical-process simulation models are based on mathematical simulations of the overland erosion process, and would provide the best estimates of soil losses and sediment yields from uranium tailings impoundments and surrounding watershed areas.

4. Survivability of Ancient Man-Made Earthen Mounds

In this study, we reviewed the construction techniques and the long-term survivability of ancient earthen mounds in the British Isles, Europe, Asia Minor, North Africa, and the United States. Evidence in the archaeological record suggests that large man-made earthen structures can remain sound and intact for time periods comparable to those required for the stabilization of tailings impoundments. Our report on this subtask presents archaeological evidence on the existence and survivability of man-made earthen and rock

structures from around the world, and reviews the factors that contributed to their survival or destruction. Our study results are summarized in the following paragraphs.

To increase mound stability, the following design features and location conditions should be either avoided or protected against:

- the use of retaining walls (timber or other materials) to maintain steep side slopes that reduce the initial earthen material requirement
- the use of steep slopes such as 1V:2H that lead to slope failures (slumping)
- the lack of rock covering (armoring), especially in semi-arid climates where it is difficult to establish vegetation, but where local rainfalls are intense
- flood plain locations where river migration is imminent
- the use of brick or brick-like materials that cannot tolerate settlement or lateral movement
- the use of materials that attract rodents, because rodents dig burrows and displace cover material
- conditions conducive to the establishment of large trees (mainly eastern USA)
- the use of mound configurations (such as pyramids) that provide corners subject to local flood scour

The design features and construction methods of the more stable mounds are listed below:

- the use of protective rock cover
- layered and compacted construction of the earthen covers
- the use of staged construction where each stage is stabilized before others are added
- designs that use interior stabilization such as radial and concentric circular walls
- the use of terraced embankments for slope stability and the provision of solid foundations for each successive stage (multiple levels)
- the use of a stable foundation such as bedrock or an excavated and compacted foundation
- the use of a vegetative cover where climate permits (eastern USA)

- an initial maintenance period and design flexibility to mitigate instabilities. This would coincide with staged construction.

Additional conclusions regarding the stability of archaeological mounds are as follows:

- tree growth seems to cause disruption only in extremely wet climates
- windblown deposits within the rock covers were not in evidence
- embankment slump failures are more common in wetter climates (eastern USA), and sheet erosion is more common in the more arid climates. The wetter climates support vegetation which would effectively prevent serious overland erosion, but excessive soil moisture combined with steep side slopes may result in slumping.

Draft Interim Riprap Handbook

Using results from the baseline data studies, a draft copy of the document Interim Handbook for the Design and Application of Riprap to Mitigate Wind and Water Erosion of the Covers of Stabilized Uranium Mill Tailings Piles, NUREG/CR-2936 (PNL-4454), was submitted to the NRC for review. A brief summary of the handbook contents follows.

In the handbook, the design of a rock cover for uranium mill tailings includes designing a minimum rock cover to resist wind and precipitation run-off erosion using the Universal Soil Loss Equation (USLE) as a basis for design. A rock cover (riprap) is specified to resist the design flood erosion (probable maximum flood) using an analytical method developed by Simons and Stevens at Colorado State University. This analytical method provides a framework for computing rock size as a function of the tractive stress on the rock particles and can be applied to any channel side slope, flow direction, and velocity.

The rock for erosion protection is chosen based on a systematic field and laboratory investigation to select the best available rock. Minimum acceptability criteria that concentrate on durability are given for the rock. The handbook specifies conventional construction methods for rock placement along with a program for quality control and inspection. The interim handbook also gives an example problem illustrating the use of the design equations.

Engineering Studies and Riprap Handbook Development

The three subtasks under this part of Task 1 were specified at the beginning of FY-83. Subtask A is a continuation of the riprap design methods and baseline studies of overland erosion. Subtask B is a continuation of the literature review on rock durability and weathering completed before this reporting period. Subtask C extends the handbook development work. The following subtask descriptions and discussions of completed work apply only to the first two quarters of FY-83. At the end of the second Quarter, FY-83, all subtasks were redefined as discussed under Project Management.

Subtask A. Technical and Economic Feasibility Study

The objectives of this task are to apply the design procedures to be presented in the handbook and evaluate whether they meet NRC's needs adequately, and to assess the technical and economic feasibility of using riprap to stabilize uranium tailings covers. To achieve these objectives we will address several issues that are important to the ultimate acceptance and use of the handbook by the uranium mill licensees. Some of the issues we will address are:

- Are the design procedures in the handbook understandable and adequately explained?
- Will adequate data typically be available to apply the handbook procedures?
- Are suitable riprap materials generally available within an economical distance of the uranium mill sites?
- What are the uncertainty levels associated with determining the key design parameters and are they acceptable?
- Will the design procedures in the handbook provide "reasonably achievable" long-term stability of tailings pile covers?

In order to accomplish the above objectives, we will select at least two tailings sites following a field reconnaissance evaluation and apply the handbook techniques. We will consider decommissioned sites because conditions representative of those to be encountered in the design of riprap can more easily be defined. Once these sites are selected, we will conduct hydrologic and hydraulic analyses to determine input parameters for the riprap design method. Typical design parameters would be mean flood-flow velocity and design flood elevation. Emphasis will be placed on utilizing available data and conventional analytical techniques because they are representative of the conditions facing either a licensee, or an NRC evaluation of the decommissioning plan.

A reconnaissance trip of tailings disposal sites was conducted during November and included visits to sites in Colorado, New Mexico, and Texas. The purpose of the trip was to select at least one or possibly two sites as test cases where we can apply and evaluate the riprap design procedures proposed in the handbook. Based on the trip results, the sites at the Slickrock Mill (Near Dove Creek, Colorado) and the Shiprock impoundment (Shiprock, New Mexico) were tentatively selected for analysis. Both are decommissioned sites and experience overland erosion and gully problems. Rock riprap was used at the Slickrock site to remedy some localized erosion and to reduce the effects of overflow from the Dolores River. A combination of river sand, gravel, cobbles, and boulders was applied as cover material to protect against erosion at the Shiprock site. The decommissioned impoundment at Grand Junction, Colorado was selected as an alternative site for study.

Subtask B. Rock Durability Study

In this study we evaluated an ultra-sonic rock-testing technique to determine its applicability for testing the long-term durability of rock riprap. Initially, we collected rocks of known approximate weathering exposures and tested them using an ultrasonic procedure. We presented the results of the laboratory testing of the rock samples in an interim draft report to the NRC. The initial results of the ultrasonic testing have largely determined the direction of our remaining work on this subtask.

During October, we collected dated rock samples from a rock glacier on Mt. Sopris near Aspen, Colorado. The samples were returned to Pacific Northwest Laboratory and tested during November and December. Attempts were made to collect other dated rock samples from archaeological sites. We made field trips to Mesa Verde National Park (Colorado) and Chaco Canyon National Park (New Mexico) in early November to determine the availability of dated rock samples. Although suitable samples exist at the sites, permission to remove the samples from the park sites was denied by the National Historic Trust and the State Historical Commission. Therefore, we reported only on the Mt. Sopris samples.

The interim report on the initial rock durability testing was sent to the NRC in early April 1983. Based on the report results, we revised the scope of the rock durability study as described in the following paragraphs.

The objective of the study was to determine a method that the NRC can use to evaluate the durability of rock proposed for riprap protection of tailings impoundments. The hypothesis that a correlation exists between ultrasonic velocity attenuation and rock strength was tested using rock samples with known approximate exposures to weathering, ranging from a few hundred to 20,000 years. We assumed that rock strength, hence ultrasound velocity, would decrease the longer the rock was exposed to weathering. Based on the test results, the following conclusions are offered:

1. There was no correlation between the velocity and attenuation measurements and the duration of weathering (10^2 - 10^4 years) in the rocks tested.
2. There appears to be a general trend for the rocks to exhibit more isotropic velocity and attenuation with increasing duration of weathering. This phenomenon may be useful for evaluating nondurable rock for rock armor applications.
3. The testing technique requires equipment and tests that are more sophisticated and elaborate than could be reasonably expected in mill operations.

The rock selection and durability studies proposed for FY-83 will be closely coordinated with the technical feasibility and economic studies. Using actual decommissioned impoundment sites selected in Subtask A, we will use geomorphic techniques to determine those outcrops of rock, and associated weathering debris, in the immediate vicinity and in progressively increasing distances from the sites that are most resistant to weathering. The relative

ages of the rock will be determined using semi-quantitative techniques, and samples will be collected for testing. The samples will be subjected to standard tests for rock durability and ranked according to test results. The samples will then be re-evaluated using the information from the engineering and economics studies. We will use this integrated approach to develop the final rock selection and testing procedures to be recommended in the handbook.

Subtask C. Development of Interim Riprap Handbook

No work was performed on this task during the quarter. The publication milestone has been cancelled, based on NRC comments, and future work will be devoted to the more comprehensive final handbook.

Project Management

During the second quarter of FY-83, we revised the subtask structure and scope based on NRC comments (revised 189) and the results of the baseline studies. We cancelled experimental work and added certain subtasks to provide a much more comprehensive handbook. The additional subtasks address embankment slope stability, the use of vegetation and earth-rock protection, and local drainage problems around impoundments. The revised subtask breakdown for FY-83 is presented in the following section.

PROJECTED WORK FOR NEXT YEAR

Subtask A. Technical Feasibility Study

The riprap design methodology to be presented in the handbook will be tested on at least two decommissioned uranium tailings impounds. We will evaluate the use of backwater profiles and flood routing techniques in determining the hydraulic design criteria. A sensitivity analysis will be performed on the hydraulic computations to evaluate the effects of parameter uncertainties on riprap size. We will evaluate and recommend methods for computing shear stress on the riprap due to flowing water and determine final design procedures for the rock riprap and filters. Also, the results of Subtask B (Overland Erosion and Local Scour) and C (Local Drainage) will be integrated into the design process.

Subtask B. Riprap Design for Overland Erosion and Local Scour

In this subtask we will determine procedures to be used to design riprap covers that will protect tailings impoundments against overland erosional processes resulting from rainfall-runoff. The erosional processes to be considered are sheet and gully erosion. Another part of the subtask will review, evaluate, and recommend design criteria to mitigate local scour from flood erosion. Flood flows passing around and/or over decommissioned tailings impoundments can cause non-uniform flow conditions that may lead to localized erosion of the earth cover. Criteria for riprap designs that reduce both overland erosion and local scour will be integrated into the riprap design procedures of Subtask A.

Subtask C. Riprap Protection for Local Drainage Channels

Some tailings impoundments may require local drainage channels or water diversion structures to collect the excessive runoff or flow from small stream channels and route it around the structure. We will review and document the local drainage problem for tailings impoundments using available data. Procedures used to design riprap for the lining and protection of earthen channels and canals will be reviewed and design procedures recommended for the handbook. We will integrate results of this subtask into the procedures for designing riprap in Subtask A.

Subtask D. Vegetation Covers

In this subtask we will develop criteria for the use of vegetative covers to protect tailings impoundments where climatic conditions permit. At impoundment locations where rock sources are very scarce, vegetation may be feasible for erosion protection if adequate moisture is available. Vegetation may also be used together with a gravel/cobble layer and rocky soil. We will review the technology available for applying vegetation as erosion protection and determine the marginal conditions in climate and soils that would limit the use of vegetation. We will investigate the use of vegetation with gravel or earth-rock material and recommend methods based on available data.

Subtask E. Economic Evaluation of Riprap Design Procedures

In this subtask we will analyze the relative costs of various riprap design conditions and geologic materials that are available for armoring the impoundment covers. The variations in design would be changes in embankment slope and impoundment shape. We will compare the relative costs of different trial designs for a series of embankment slopes to evaluate cover materials such as quarry-run stone, river gravel and cobbles, rock debris (talus), and others. The work in this subtask will be closely coordinated with Subtask A (Technical Feasibility) and Subtask G (Rock Selection and Durability).

Subtask F. Watershed Model Evaluation

In this subtask we will evaluate the use of watershed models to determine the long-term erosion of the watershed surface immediately surrounding the impoundment and the earthen radon barrier. The models will be evaluated on their ability to determine erosion rates from single rainfall events, annual long-term erosion rates, scour and deposition areas of the watershed, and possibly, the potential for gully migration.

Subtask G. Rock Selection and Durability

The work in this subtask will involve field investigations to determine rock sources, types of geologic material available, and the verification of field procedures for rock selection. We will select at least one of the Subtask A sites and identify outcrops of potential cover-armor rock and associated weathering debris in the immediate vicinity of the site and in progressively increasing radii from the site. We will evaluate the relative ages of the rock using accepted geomorphologic techniques and collect samples

for testing. The most resistant rocks will be ranked and then re-evaluated using the information from Subtask A and E. This integrated approach is similar to that which would be used by mill operators, and will provide the rock selection and testing procedures for the handbook.

Using the above results, the baseline literature review, and other sources of data on rock weathering and durability, we will investigate the feasibility of providing estimates of weathering rates. The weathering rates would be used as design criteria to estimate the reduction in individual rock size and weight over long periods of time.

Subtask H. Impoundment Slope Stability and Settlement

We will review literature that documents the problems of embankment stability and settlement in tailings impoundments. We will review the soil mechanics techniques that are used to analyze slope stability and differential settlement and recommend criteria to evaluate the techniques for the final handbook. The criteria would be used primarily to analyze embankment slope stability because unstable slopes can easily lead to failure of the cover protection.

TASK 2. INTERIM STABILIZATION

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

FIN B-2370 (Task 2)

OBJECTIVE

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

ACCOMPLISHMENTS DURING PAST YEAR

Subtask A. Laboratory Studies

The major activities of this task are to identify, and evaluate under controlled laboratory conditions, the materials commercially available that are applicable to the control of fugitive dust from uranium tailings piles. A technology review, including both literature searches and contacts with manufacturers and users of dust control products, has led to the identification of more than 40 chemical stabilizers and several physical stabilizer systems. Examples of physical stabilizers are woven and non-woven mats or geotextiles and newly developed windscreen systems.

We conducted laboratory tests to evaluate the chemical stabilizers and to select the more effective ones for field testing on a uranium tailings pile. The laboratory studies consisted primarily of wind erosion tests performed in a wind tunnel. Samples of treated simulated tailings were tested at different stabilizer dilution and application rates and under various accelerated weathering conditions. These weathering tests included freeze/thaw cycling, wet/dry cycling, and intense ultraviolet light exposure. In addition, we conducted laboratory tests to compare the stabilizers' resistance to water erosion, their effect on the hydraulic conductivity of the tailings, and effects of the tailings and/or dilution water chemistry on the effectiveness of the stabilizers.

Subtask B. Field Studies

We selected fifteen chemical stabilizers for field testing based on the results of the laboratory tests. After considering several possible test sites, the Federal American Partners uranium mill in Gas Hills, Wyoming was chosen as the location for the field test. The field test was initiated in August 1982. We prepared the test area by removing the top layer of tailings, which had previously been treated with a chemical stabilizer by the mill operators. The site was then leveled and compacted by discing and harrowing the test area.

Fifteen stabilizers were applied to the tailings at two different application rates, for a total of 30 test plots. Fourteen of the materials were liquid chemical stabilizers and were applied with a small field sprayer that was modified for this purpose. The other stabilizer, a wood fiber mulch, was applied as a water slurry and required a "Hydroseeder," a tank with a high pressure pump and nozzle, for application.

Since application of the stabilizers in August 1982, we have periodically visited and visually inspected the field test site. Information from these monitoring trips is documented by photography, and is supplemented by meteorological data that are automatically collected by a portable weather station and data logger located on the field test site. We use the weather data to correlate weathering conditions with our observations of degradation of the stabilizers over time.

PROJECTED WORK FOR NEXT YEAR

We are planning to augment the 1982 field test with additional field testing of stabilizer systems that will be applied during the summer 1983. New and promising chemical stabilizers that have recently been identified may be tested at that time. In addition, we are investigating a physical method of fugitive dust control (the use of windscreens) for potential use on uranium mill tailings. Windscreens have been used extensively for snow drift control and other applications. However, recent innovations in this technology indicate that some systems may be effective as a means of reducing or eliminating the fugitive dust generated from the surface of tailings piles. A field test of these dust control windscreens is planned for this summer. We will continue to periodically monitor the site and collect weather data during the next year to evaluate all the stabilization methods being tested at both field sites.

Information from the laboratory and field testing of the stabilizer systems, cost data, and other available information will be assembled into two topical reports. The final monitoring and evaluation report will describe the engineering specifications for the use of current technology for fugitive-dust control that applies to the interim stabilization of uranium mill tailings.

TASK 3. TAILINGS DEWATERING TECHNIQUES

Task Project Manager: DW Mayer
Principal Investigator: DW Mayer
Support Investigators: TJ McKeon
SW Tyler

FIN B-2370 (Task 3)

OBJECTIVE

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

ACCOMPLISHMENTS DURING PAST YEAR

Subtask A. Evaluation of Linear Consolidation Theory

The objective of this subtask is to evaluate the classical vertical consolidation theory, as used in the TRUST variably-saturated flow code, and determine its applicability for estimating the settlement of wet tailings sluiced into burial pits. In addition, special consideration will be given to the determination of the best measurement techniques for obtaining the required model input values in the laboratory.

The research performed last year has shown that the consolidation theory utilized in the TRUST code is inadequate. Specifically, the classical consolidation theory (proposed by Terzaghi in 1925) which is based on the concept of effective stress (the total stress minus the pore-water pressure), is only valid in the saturated zone. In an attempt to extend this theory into the unsaturated zone, Bishop (1959) introduced the Chi factor concept which is a correction to the pore-water pressure term in the effective-stress equation. The Chi factor approach has been shown to be inadequate when the soil is below a certain level of saturation. In particular, the critical saturations for clays and clayey silts is 0.85, for silts and fine sands it is 0.50, and for sands the critical saturation is 0.20. The tailings are likely to be below these critical levels of saturation much of the time, and therefore, these findings indicate that a better theory is required.

Recent work by Fredlund and Morgenstern (1976) shows that the stress state in an unsaturated medium should be described as a function of two stress-state variables rather than one stress-state variable such as the effective stress. The variables selected by Fredlund and Morgenstern are the total stress minus the pore-air pressure, and the pore-water pressure minus the pore-air

pressure. Based on these findings, the most accurate method for describing consolidation in an unsaturated medium such as uranium mill tailings would be the two stress-state approach proposed by Fredlund and Morgenstern. These results have been discussed in more detail in a report that was submitted in August, 1982.

We also reviewed the literature that pertains to the acquisition of data on saturation, hydraulic conductivity, and void ratio as a function of the two stress-state variables. Based on our findings we submitted a letter report (as an addendum to the fourth quarter 1982 report) that discusses the techniques used to obtain the required data.

Subtask B. Measurement and Representation of Material Characteristics

Our objective in this subtask is to obtain the saturation, hydraulic conductivity, and void ratio data that are necessary to perform simulations of the fluid flow and consolidation in uranium tailings piles. We will also develop the methods to interpret the lab data.

Based on the findings from Subtask A, we obtained the necessary equipment to measure the soil characteristics. This equipment consists of a consolidation apparatus that is used for obtaining the data for saturated samples, and a tri-axial test set that is used for measuring the soil characteristics of unsaturated samples. In addition, methods for representing the soil properties as mathematical functions have been developed and a method for estimating hydraulic conductivities of unsaturated soils based on the saturation data has been implemented. It is often necessary to estimate the hydraulic conductivities for the unsaturated zone because this property is difficult to measure.

Subtask C. Computer Code Modification Testing and Documentation

The objective of this subtask is to change the existing consolidation model in the TRUST code to a model incorporating the two stress-state variables. In addition, the new version of TRUST (which will be designated TRUST-II) and the pre- and post-processor utility package will be tested and documented.

We have completed the major modifications to the TRUST-II code and initial testing is under way. Preliminary results indicated that in addition to the void ratio, the saturation and hydraulic conductivity properties should be made a function of the two stress-state variables. This modification has also been incorporated. As soon as data are available from Subtask B, we will begin additional testing of the model.

The TRUST-II utility package consists of several programs that are useful for preparing input data for TRUST-II and for analyzing the results of the TRUST-II simulations. The utility package consists of a grid generation program, programs for analyzing the soil property data, and a program that is useful for analyzing the flow and transport of contaminants. This package has

been successfully tested and the documentation is essentially completed. We will submit a report on this package in August.

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

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

Subtask E. Tailings Dewatering Test Cases

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

PROJECTED WORK FOR NEXT YEAR

The work to be performed during the remainder of FY83 and for FY84 falls into three main categories. First, we will determine the soil properties that affect the flow of fluid and the consolidation of uranium mill tailings. Second, we will test and document the TRUST-II computer model for variably-saturated flow and consolidation. Finally, the model will be used to perform several comparison studies and a demonstration case will be run.

To measure soil properties relevant to the study, we must acquire saturations, void ratios, and hydraulic conductivities of typical soil and tailings samples from a uranium mill tailings disposal site. These properties must be measured in both saturated and unsaturated samples as functions of the two stress-state variables (saturations, of course, will only be measured for the unsaturated samples). These data are necessary input to the model so that flow and consolidation can be accurately simulated.

The TRUST-II model will be tested and documented to insure that the new theory incorporated into the model is working properly, and that it does not adversely affect the accuracy of the model results when the consolidation portion of the model is not being used. Once documented, the model will be available for use by others. Note that the utility package that we will submit in August 1983 is compatible with only the TRUST-II version of TRUST. This makes the testing and documentation of TRUST-II an important goal for FY84, because the utility package will not be compatible with the currently available version of TRUST. Our decision to make the utility package compatible with TRUST-II eliminates the need for redocumentation of the utility package when the TRUST-II model is released.

When the measurements phase of this task is complete and when the model has been suitably tested, we will use the model in several comparison studies. These studies will determine the effect on model results caused by using the classical consolidation theory or the new, two stress-state theory that has been incorporated into the model. Additionally, we will undertake studies to determine if the hydraulic conductivity must be allowed to vary with void ratio. If significant consolidation occurs, then the void ratio may change enough to have a significant effect on the flow results.

Our major accomplishment to be achieved during FY84 will be demonstrating the capability of the model on a typical uranium disposal site. This study will use the soil and tailings data as input to a simulation to evaluate the flow and consolidation at a typical site, and will thereby demonstrate the use of this analysis technique. The result will be a proven technology for the analysis of tailings dewatering in disposal pits.

Although this will complete the planned work for FY84, a logical follow-on study suggests itself. The analysis capability developed under this task will be the best available and as such is quite sophisticated. Certain simplifications may possibly be acceptable. To determine the minimum acceptable analysis, an accurate simulation of an actual test site should be performed. The results from this study would serve as a reference, against which we could compare simplified analysis techniques. By analyzing the results from the simplified techniques with the reference case, it would be possible to determine if and when a simplified technique was unacceptable. The results of this follow-on effort would therefore provide needed guidance on the amount of sampling and measurement of soil parameters that are necessary, and delineate the detail necessary in site characterization to obtain reasonable estimates of the water flow and consolidation that could be expected at a disposal site.

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

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FIN B-2370 (Task 4)

OBJECTIVE

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

ACCOMPLISHMENTS DURING REPORT PERIOD

Subtask A. Neutralization Methods Selection

As a first step in assessing the effectiveness, benefits and costs of tailings and tailings solution treatment, we conducted a review of existing literature on tailings amelioration techniques. We also assessed the effectiveness of such techniques in reducing contaminant mobility.

Three general amelioration methods were identified: neutralization, fixation, and specific constituent removal. During neutralization, a reagent is added to the tailings solution to neutralize the acidity (raise the pH), thus reducing the solubility of various pH-sensitive contaminants. In contrast, fixation processes add materials such as lime, cement or asphalt to the waste to produce a physically stable composition that resists leaching of hazardous constituents. Specific constituent removal encompasses varying techniques, such as alternate ore-leaching processes, effluent treatment with sorption, or ion exchange agents or selective precipitation that reduce specific constituent concentrations in tailings solution.

At present, neutralization processes appear to be best suited for treating uranium mill tailings because they can limit the solution concentration of many contaminants at a reasonable cost, and thus reduce the potential for ground-water contamination. However, the effectiveness of the process depends on the reagent used as well as the waste being treated. Of the six reagents being studied (lime, limestone, caustic soda, soda ash, combined limestone/lime, and combined alumina/lime/soda), a combined treatment of limestone and lime seems best. The combined treatment works especially well for tailings

containing ferric iron because the limestone economically buffers the solution acidity, while the lime takes the pH to ~8.0, an optimum level for heavy metal removal. For those tailings containing ferrous iron, lime alone works best. In general, the costs for the lime/limestone or lime processes range from \$0.20 to \$1.00 per 1000 gallons of treated water, excluding capital equipment costs.

Because the above conclusions are based upon results from the neutralization of acid mine drainage and Canadian uranium mill wastes, future work in this area will include controlled laboratory experiments comparing various neutralizing agents for treatment of acidic uranium mill tailings solution from representative U.S. sites. Each reagent will be evaluated in terms of:

- the quality of treated effluent (with greater detail on radionuclides, potentially toxic trace metals and mobile anions)
- the properties of the neutralized sludge (e.g., dewaterability, hydraulic conductivity)
- the cost of the reagents for typical U.S. uranium mill tailings treatment.

In general, fixation processes are not currently studied for treatment of uranium mill tailings because they substantially increase the volume and weight of waste to be disposed, are very expensive (\$1.5 to \$105 per ton of tailings) and often are not effective in providing complete containment of potentially hazardous constituents after tens of years. On an equivalent cost basis (cost per 1000 gallons of waste) and assuming a waste-slurry specific gravity of 1.4, solidification/fixation would cost from \$8.7 to over \$400 per 1000 gallons. Of the three types of solidification processes, cement-based ones are least expensive at \$1.5 to \$7 per ton of waste, lime-based processes cost \$10 to \$17 per ton, and asphalt-based processes cost \$120 to \$420 per ton. Currently, any application of solidification/fixation processes to disposal of U.S. acidic mill tailings would need to be limited to only a fraction of the total waste stream such as a concentrated portion of highly hazardous material. As an example, barium/radium sulfate sludge might be a candidate for solidification if the U.S. mills should initiate barium chloride (BaCl_2) treatment to remove radium (^{226}Ra).

To date, the only constituent for which specific removal is practiced is ^{226}Ra . Specific radium removal is practiced at all currently operating Canadian mills. The chosen ^{226}Ra treatment process is BaCl_2 addition to neutralized effluent streams. The BaCl_2 in the presence of high sulfate concentrations causes BaSO_4 to precipitate and scavenge (co-precipitate) $(\text{Ba},\text{Ra})\text{SO}_4$. If the waters are initially low in sulfate, Na_2SO_4 may be added to promote precipitation. The effluent is treated in stirred reactors and settling of the precipitate is enhanced by addition of FeCl_3 and organic polymers within flocculation tanks. The supernate water from flocculators is often further clarified on inclined plate clarifiers or filtered through packed beds of anthracite coal and sand. The dissolved ^{226}Ra is readily reduced to the current Canadian concentration guide of 3 pCi/l, but

reduction of the total radium concentration (dissolved plus suspended) to 10 pCi/l is not readily met. The $(\text{Ba,Ra})\text{SO}_4$ precipitate is rather fine-grained and is not readily removed from solution without large settling basins and long residence times. The Canadian mill operators suggest that 30 pCi/l for a total-radium radiation protection concentration guide (as currently exists in the U.S.) would be attainable. Capital costs for stirred reactors, settling ponds and sand filters for a 1000 gallon/minute radium removal plant run \$25,000 (1978 dollars) and operating costs were \$0.68/1000 gallons (i.e., within the range of current neutralization costs).

We also reviewed ion-exchange removal of radium because it is used to remove radium from low quality municipal and well drinking-water sources and mine waste waters. Common cation exchange resins such as Dowex AG50W are of limited use on the high acid/high salt tailings solutions because calcium, magnesium, and other multivalent cations load the exchange sites and compete too strongly. Other specialized resins such as Chelex 100, manganese impregnated acrylic fiber resins, and Decalso (a synthetic zeolite) show good radium selectivity from neutral mine waters with low total dissolved solids (TDS), but column regeneration is a problem. Without good column regeneration the cost for treatment is high. The most promising ion exchange resin is a new cross-linked organic polymer XF-43230 (Dow Chemical) recently studied by Boyce and Boom (1982). Using a neutral-pH mine water with ~1350 ppm TDS and up to 100 pCi/l ^{226}Ra , the new resin removed >95 percent of the ^{226}Ra for seven months, while a standard, strong-acid cation exchanger lasted only one week before calcium loading decreased radium removal. At best, ion exchange appears useful for removing radium only from neutralized, low-TDS waters. Direct application of ion exchange resins to acidic tailings liquids with 15,000 to 35,000 ppm TDS appears inappropriate. No specific costs for ion-exchange removal of radium for mine waters are available, but for a 1000 gallon/min drinking water plant using conventional strong-acid cation resins, the capital costs are estimated at \$1,000,000 (1976 dollars) and \$0.20/1000 gallons for operating costs. The capital costs greatly exceed those for BaCl_2 treatment, but the operating costs are 3.5 times less.

The lime/limestone neutralization process is sufficient to reduce ^{230}Th , ^{210}Pb , ^{210}Po and many pH sensitive trace metals in tailings effluent liquids to acceptable levels so that further treatment is not warranted according to several Canadian authors (Moffett 1976; Itkovitch and Ritcey 1979; Raicevic 1980).

There have been no attempts to specifically remove constituents such as SO_4^{2-} , NO_3^- and NH_4^+ from the effluents from neutralized tailings in Canada nor from acid effluents from U.S. tailings, even a though they exceed concentration guidelines. Classical removal techniques for these three constituents (sulfate by BaCl_2 treatment and anion exchange, nitrate by anion exchange or denitrification, and ammonia by stripping, nitrification, or cation exchange) were reviewed, but applicability and costs for treating acidic uranium mill tailings are unknown.

We reviewed novel ore-leaching or tailings-leaching processes that remove constituents before disposal. In general, new ore processing techniques (e.g.,

HCl or HNO₃ leaching) do not appear to offer significant cost advantages and they also generate potentially hazardous waste streams. Tailings extraction techniques that selectively remove ²²⁶Ra but leave behind ²³⁰Th are not useful over long time periods (10³ to 10⁴ years) because the ²²⁶Ra concentration may return to previous levels as the ²³⁰Th decays.

To evaluate the most economical and practical method of assuring long term safety of uranium mill tailings disposal options one must consider the natural ability of soils and sediments to neutralize acid wastes and to retard the migration of hazardous constituents via adsorption and precipitation interactions. Field observations (e.g., Cherry et al. 1982; Dodds 1979) and laboratory results (Gee et al. 1980) suggest that contaminant transport around tailings pits is limited for pH sensitive metals and radionuclides as long as the solution pH remains above pH 4. Moffet (1976) reviewed the hazard potential of neutralized tailings and concluded that ²²⁶Ra represented the largest health hazard and nitrate, sulfate and ammonium also could leach from tailings at concentrations significantly above water concentration guides.

These observations lead us to conclude that the most mobile constituents in uranium tailings effluents will probably be the anions SO₄²⁻ and SO₄³⁻ both of which can exceed drinking water and irrigation/livestock concentration limits by several orders of magnitude.

Subtask B. Laboratory Analysis

Laboratory experiments were conducted to evaluate the performance of selected neutralizing agents for the treatment of uranium tailings solutions. Highly acidic tailings solutions (pH<2) from the Lucky Mc Pathfinder Mill in Gas Hills, Wyoming and the Exxon Highlands Mill near Casper, Wyoming were neutralized to a pH of 7 or greater using seven neutralizing agents. Reagents used included:

1. Fly Ash from Boardman Coal Plant, Boardman, Oregon
2. Fly Ash from Wyodak Coal Plant, Gillette, Wyoming
3. Calcium carbonate (CaCO₃) reagent grade
4. Calcium hydroxide [Ca(OH)₂] reagent grade
5. Magnesium oxide (MgO) reagent grade
6. Sodium carbonate (Na₂CO₃) reagent grade
7. Sodium hydroxide (NaOH) reagent grade.

We evaluated the effectiveness of the selected neutralizing agents in treating uranium tailings solutions under controlled laboratory conditions based on three criteria. The criteria are: 1) treated effluent water quality, 2) neutralized sludge handling and hydraulic properties, and 3) reagent costs and acid neutralizing efficiency.

Treated effluent water quality suggests that the removal of many dissolved constituents (e.g., Al, Fe, Si, As, Ag, Cr, Cu, Pb, V, Zn, ²¹⁰Pb, and ²³⁰Th) is controlled by pH buffering and does not depend upon the specific neutralizing agent. However, test results show that effluent concentrations of sulfate, cobalt, ²³⁸U, and ²²⁶Ra depend on the reagent selected. Only those reagents

containing calcium which promotes the formation of gypsum removed sulfate from solution. Cobalt, ^{238}U , and ^{226}Ra are not removed from solution as effectively with carbonate reagents as with other reagents. This effect may be a result of the formation of soluble carbonate complexes of cobalt, uranium, and radium. Other constituents (e.g., K, Na, Ca, NO_3 , Cl, and Se) show little or no change in solution concentration during neutralization regardless of the reagent used. Removal of manganese and molybdenum is variable and does not seem to correlate with the reagent used.

The neutralized Lucky Mc Pathfinder tailings solutions exceed EPA's maximum concentration levels: by 10^2 to 10^3 times for Se and Mn; by 10^1 to 10^2 times for Mo, SO_4 , and TDS; and by 10^0 to 10^1 times for NO_3 , Cl, and ^{226}Ra . The Exxon Highland Mill tailings solution after neutralization exceeds maximum concentration levels; by 10^1 to 10^2 for Se, Mn, and SO_4 ; and by 10^0 to 10^1 times for Cl, TDS, and ^{226}Ra . Thus, one might conclude that potentially, the most problematic constituents will be Se, Mn, SO_4 , and total dissolved solids. Most likely, more Mn would be removed upon contact with sediments, but Se, SO_4 , and TDS would require significant dilution with low ionic-strength ground water or specific treatment to ensure that their concentrations are below water quality guidelines.

We also used selected sludge properties to evaluate the neutralizing agents. We determined the total weight of sludge generated, moisture retention of the sludge, and ran mineralogical analyses of the sludge for each of the reagents tested. Results of the sludge characterization indicate that calcium-containing reagents and fly ash produce the largest amount of sludge. Reagents without calcium produce much less sludge, but of a substantially higher moisture content. Crystalline mineralogy of the two fly ash samples compared to those reacted with tailings solution indicated that lime (CaO) and periclase (MgO) present in the ash were dissolved during contact. Gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) was identified as the only crystalline phase detected in the sludges neutralized with calcium reagent. We did not detect crystalline phases in samples neutralized with magnesium oxide, sodium carbonate, or sodium hydroxide. Undoubtedly all sludges contain noncrystalline precipitates that form rapidly during neutralization.

We monitored the amount of reagent required to neutralize the tailings solution to pH ~ 7 to determine the efficiency of the reagent and costs. Information on the use of reagents indicates that the quantity of reagent required to treat a given volume or solution depends on the total acidity of the solution, the equivalent weight of the reagent, and the efficiency with which that reagent reacts to neutralize solution acidity. Results of the reagent usage tests indicate that a high-quantity total-acidity tailings solution (e.g., Lucky Mc Pathfinder 15,000 mg/l total acidity) may require as much as three times the amount of reagent required to neutralize a low-quantity, low-quality leachate (e.g., Exxon Highland Mill 4500 mg/l total acidity). Results of tests for neutralization efficiency suggest that only two of the reagents tested had efficiencies in excess of 90 percent of their theoretical neutralization capacity (e.g., sodium hydroxide = 96 percent and calcium hydroxide = 90 percent). Calcium carbonate exhibited the lowest neutralizing efficiency of the reagents tested (67 percent). No efficiency could be calculated for

the fly ash samples because they have no known equivalent weight. Table 1 gives the costs of reagents and shows that calcium carbonate, calcium hydroxide, and magnesium oxide are the least expensive of the neutralizing agents tested. The reagent costs for the Lucky Mc Pathfinder solutions (high acidity) calculated per ton of ore processed for treatment with calcium carbonate, calcium hydroxide, and magnesium oxide were \$0.88, \$0.83, and \$0.77, respectively, while the costs for the Exxon solution were \$0.30, \$0.26, and \$0.26, respectively. Costs for other reagents tested were at least twice as much. Sodium hydroxide was the most expensive reagent examined with a reagent cost of nearly ten times that of calcium carbonate, calcium hydroxide, or magnesium oxide.

TABLE 1. Treatment Costs for Exxon and Lucky Mc Pathfinder Solutions^(a)

Sample Number	Reagents	Exxon	Lucky Mc Pathfinder
1	Fly Ash (Boardman)	\$0.61	1.43
2	Fly Ash (Wyodak)	0.89	1.58
3	Limestone	0.30	0.88
4	Lime	0.26	0.83
5	Magnesia	0.26	0.77
6	Soda Ash	1.01	3.54
7	Soda Caustic	2.42	7.65

(a) Reagent cost per ton of ore processed. Assumes 50-gal solution/ton of ore processed.

On the basis of these laboratory results, either calcium hydroxide or its dehydrated form, CaO (lime), appears to be the most effective option for treatment of uranium tailings solutions. In addition, cost estimates for adding the neutralization step in the overall uranium production and waste disposal scheme range from about 1 to 5 percent of the production costs. However, more sophisticated cost estimates should be performed to verify these calculations.

We performed laboratory studies to investigate the optimum pH level for neutralization of acidic tailings solution. The optimum pH is defined as that pH at which addition of reagents no longer significantly increases contaminant removal. Our results, which are on initial analyses of macrocation and anion concentrations and of the relative volume of sludge generated by further addition of a neutralizing agent, indicate that neutralization to pH ~7.2 removes the highest percentage of constituents.

We began additional experiments this year to further evaluate the two most promising neutralizing agents, hydrated lime $[\text{Ca}(\text{OH})_2]$ and limestone, CaCO_3 . These studies will evaluate the effectiveness of a two-component neutralization scheme that uses lime and limestone together to remove contaminants. The effectiveness of the two-component scheme will be determined based on effluent water quality, cost, and sludge generation and handling properties. Through our literature review, we found that a two-component neutralization scheme was cost effective when used to treat acid mine drainage. If the requirements for disposal of tailings and tailings solution include neutralization, similar cost-savings may be possible in the uranium industry.

Subtask C. Field Tests

We submitted a plan for a field demonstration which will evaluate the effectiveness, costs, and benefits of neutralization of acidic uranium mill tailings solutions to reduce the potential leaching of toxic trace elements, radionuclides, and macro ions from a tailings impoundment.

The effectiveness of the neutralization of acidic uranium mill tailings solution on a field scale will be judged on the following four criteria: 1) treated effluent water quality, 2) reagent costs and acid neutralizing efficiency, 3) neutralized sludge handling and hydraulic properties, and 4) long-term contaminant immobilization.

The plan proposes three separate alternatives to evaluate the selected neutralizing reagents or techniques in the field. The three alternatives, only one of which would be implemented, are designed to assess how the contaminants are immobilized when acid solution is neutralized.

To neutralize uranium mill tailings solution we must add an intermediate step between the exit point of the acid solution/slurry from the milling process and its discharge into an evaporation or drainage pond. During the intermediate step, the acid tailings solution will react with a neutralizing reagent(s) in a large capacity mixing apparatus until a preset solution pH (determined by laboratory experiments as most desirable) is reached. Then the slurry is discharged into the disposal pond. The primary objective of this demonstration plan is to represent the neutralization of acidic solutions in a full-scale disposal scenario as opposed to smaller-scale laboratory studies. The following alternatives describe the activities necessary to represent the field-scale treatment of acidic tailings solution.

Alternative A

Purchase outright the necessary equipment to neutralize a specified volume ($\sim 10^6$ liters) of tailings solution by using one reagent or a combination of reagents which we determine through laboratory experiments to be the most beneficial for the selected demonstration site.

Alternative B

Use the existing equipment and facilities at a milling site which currently neutralizes the acidic solution to incorporate our chosen reagent or a combination of reagents which we determine by laboratory experiments to be the most beneficial.

Alternative C

Analyze and evaluate the efficiency, costs, and benefits of an existing neutralization technique and reagent(s) in use at a mill site which currently treats acid tailings solutions.

The criteria involved in selecting one of these three alternatives are based primarily on: 1) sponsor concurrence, 2) mill site owner/operator concurrence, 3) allocation time and availability of funding for project completion, and 4) site location and equipment availability.

The field-test program is proposed to begin during the first quarter of FY-85 and will be completed during the fourth quarter of FY-87.

PROJECTED WORK FOR NEXT YEAR

Based on anticipated funding levels for Task 4 in FY-84, our efforts will focus on ongoing laboratory experiments in order to prepare a final report on tailings neutralization.

In current laboratory experiments, we are evaluating the long-term migration potential of contaminants leached by groundwater from both acid tailings and neutralized tailings sludges. The results of these studies will indicate the effectiveness of neutralization in controlling contaminant migration. We are also studying the use of a neutralizing barrier in tailings impoundments. The use of a neutralizing layer could result in significant cost savings. Only the solution that escapes the designated disposal impoundment poses an immediate contamination threat; but if a neutralizing barrier is employed, the solution is neutralized before significant migration of the solution occurs.

We will incorporate the results of these studies in a final report to be submitted in the fourth quarter of FY-84.

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TASK 5. DISPOSAL TECHNIQUES ALLOWING CONTACT WITH GROUND WATER

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FIN B-2370 (Task 5)

OBJECTIVE

Within this task we are evaluating particular environmental concerns that may result from disposal of uranium mill tailings below the water table. Disposal below the water table may be of two major types. The first involves below-grade disposal in excavated pits where a significant part of the tailings is below the regional water table and is subject to long-term leaching by ground water. The second type involves deeper disposal of tailings in underground mine stopes below the water table.

ACCOMPLISHMENTS DURING THE PAST YEAR

We studied the first type of below-grade disposal where tailings are in pits below the water table in some detail during the past year, and have completed this portion of the task. In these studies, we used models to simulate ground-water flow and transport through and away from the pits. Specifically, we simulated and analyzed cases of tailings disposal in below-grade pits above the water table and compared these results with similar simulated cases for disposal below the water table. Thus, we used already-accumulated experience for pit disposal above the water table to give us a better understanding of disposal below the water table, of leachate losses to the environment, and of the benefits of various leachate-control methods.

Three cases of pit disposal above a lateral water-table gradient were considered. A fourth case, in which the disposal pit was located above a horizontal or static water table, was considered as an index to previous studies in this area. A different leachate control method was used for each of the first three cases. In the first, or reference case, saturated sluiced tailings were in an unlined pit above the water table. The second case involved the same pit, with a clay bottom liner added. In the third case, stub-sidewall clay liners and drains in the tailings above the bottom clay liner provided still more leachate control, as did pumping the drainage effluent from the pit.

The volume of tailings leachate entering the ground water from the pits above the water table was 100 percent, 97.2 percent, and 20.9 percent, respectively, for the reference unlined pit, for the pit with the clay-lined bottom, and for the pit with the clay-lined bottom and stub-sidewalls, and with pumps to collect drainage effluent. The 75 to 80 percent decrease in leachate that

enters the ground water in the third case is significant and results from pumping the effluent from the drain sumps.

The corresponding leachate losses to ground water for the below-the-water table cases were 142.7 percent, 135.6 percent, and essentially 0 percent, based on the 100 percent loss for the reference case. Some of the tailings pore solution in this reference case is retained by capillary forces. The greater leachate losses to ground water for the first two cases below the water table (142.7 percent and 135.6 percent compared to 100 percent and 97.2 percent, respectively) are a result of the ground water flowing through the tailings. The ground water effectively sweeps all the initial pore solution from the tailings. We must remember that these leachate losses represent the leachate volume that is physically swept from the tailings by the ground water, and do not include the longer-term leaching associated with chemical interactions between ground water and tailings. Without experimental analyses of these chemical interactions, we must assume worst-case situations in the simulations to assess the overall consequences of leachate loss to the environment. Growing evidence suggests that the predicted detrimental consequences are often a direct result of the worst-case assumptions.

On the other hand, essentially no leachate was lost to ground water when the leachate control methods were combined in the below-grade disposal pit. This combination, however, requires that as the ground water enters the tailings and reaches the drains, extra water be pumped from drains. In fact, a 289.7 percent volume of fluid (about 2.9 times that of the reference case) must be pumped from the pit drains to assure that leachate will not be lost to ground water. In all of the cases where ground water is pumped from drain sumps, the cost of pumping over time periods of 1 to 10 years is a factor. Also, if the pumped fluid is not reused as process makeup solution in the milling process, it must be disposed of in an environmentally safe manner.

This study supports our earlier work concerning the effectiveness of pumping groundwater from drains in disposal pits above the water table in order to reduce leachate seepage from tailings into ground water. We also found that pumping drainage effluent from tailings drains in disposal pits below the water table controls leachate loss even more effectively (essentially no leachate loss to ground water, compared to 20.9 percent loss from pits above the water table) because greater head drawdown is possible under saturated flow conditions.

The best control of leachate from uranium mill tailings in disposal pits is provided by a combination of a clay bottom liner, clay stub-sidewall liners, and tailings drains that collect effluent for subsequent pumping from the pit. As much as 80 percent of the total leachate volume that otherwise would enter ground water from disposal pits above the water table is intercepted by this combination of control features. Model simulations of pits below the water table equipped with the same combination of controls showed essentially no leachate loss. Pumping of the effluent, however, involves continuing energy costs and requires an alternative method of disposal of any leachate that cannot be reused as mill process makeup water.

When no drains or effluent pumping are involved, the clay bottom liners are less useful. They reduce the leachate outflow rates, but the leachate continues to flow out over a much longer period of time, resulting in essentially the same total outflow volume of leachate. This pattern is about the same for disposal both above and below the water table, although more leachate enters the ground water from disposal pits below the water table.

We presented the complete results of our simulation of the fate of leachates in the NUREG/CR-3078 entitled "Model Evaluation of Seepage from Uranium Tailings Disposal Above and Below the Water Table" published in March 1983. Our conclusions and recommendations provide moderately definitive results.

PROJECTED WORK FOR NEXT YEAR

Work planned for completion during the first quarter of FY-84 will assess particular environmental concerns that may arise from the disposal of uranium tailings in abandoned deep mine stopes.

Ongoing work includes obtaining ground water, tailings, and deep-aquifer samples from two sites (one in Wyoming and the other in New Mexico), and beginning the laboratory column studies on these samples. These column studies will measure both the transport and the chemical interactions of tailings leachate with the deeper, coarse-grained aquifers. Samples of ground water, tailings, and deep aquifer materials obtained at the -700 ft depth in a Wyoming uranium mine and at the -1700 ft depth in New Mexico will be used to carry out the column experiments on migration of tailings leachate in coarse aquifer materials. Special emphasis is being placed upon leachate-aquifer material interaction in these coarse materials. In the past, coarse materials have not been studied as thoroughly as fine materials with higher clay content. Environmental problems are more likely to occur in the coarse materials because coarse materials have much lower buffering and sorption capacities than fine materials.

The experimental column work is being done cooperatively with help from NRC project B-2292 (Leachate Assessment) and from Task 4 (Tailings Neutralization) of this project. We are using inverse methods to obtain the appropriate dispersion parameters. The classical dispersion coefficient and the stochastic-covariance characterization of dispersion for the short laboratory columns are essentially identical. Accordingly, the parameter determination methods of Van Genuchten are being used to determine dispersion coefficients, which are then readily converted to the more useful stochastic-dispersion characteristics.

The initial fluid-flow analysis has emphasized the need and importance of a more complete analysis of the flow system than originally contemplated. Our original plan was to analyze situations where the natural ground-water gradient, and hence flow was directly through or at right angles to the center line of the tailings-filled stope. We have extended this to consider the three-dimensional ground-water flow through oblique cross-sections through the stopes. When ground-water seeps through oblique cross-section or more specifically along the longitudinal axis of the stope, a larger volume of tailings can be

leached, hence larger (i.e., at least deeper), contamination plumes can result. In general, limited data on aquifer hydraulic conductivity are available. Accordingly, the flow analysis is being idealized consistent with the limited data, which results in the use of analytical, multi-dimensional, ground-water flow expressions. The velocity field of the ground-water system will be obtained from these analytical expressions, and the convective pathlines obtained through simultaneous solution of the three characteristic differential equations. The latter will be solved using a modified fourth order Runge-Kutta numerical scheme. The chemical interactions determined by column experiments will be used with the pathlines to determine the characteristics of the contamination that might be intercepted if a deep domestic-water well were drilled down gradient from the tailings-filled stope.

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

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FIN B-2370 (Task 6)

OBJECTIVE

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

ACCOMPLISHMENTS DURING PAST YEAR

Subtask A. Field Comparison of Radon Progeny Measurement Techniques,
(J. A. Young)

Objective

In this subtask we ascertain whether a satisfactory, short-term measurement technique is available for the determination of annual average radon-daughter concentrations in buildings.

Accomplishments During Past Year

The Alpha Nuclear Corporation radon-progeny monitors have been delivered, but the software necessary to calculate radon-daughter concentrations from counting data have not. Therefore, continuous side-by-side comparisons of radon and radon-daughter measuring devices in two test houses in Edgemont, South Dakota have not begun.

The report Comparison of Five-Minute Radon-Daughter Measurements with Long-Term Radon and Radon-Daughter Measurements (NUREG/CR-3106, PNL-4591) has been published. This report compares the standard deviations between average annual indoor radon-daughter concentrations obtained from six 100-hour Radon Progeny Integrating Sampling Unit (RPISU) measurements and 1) single air filter measurements; 2) single RPISU measurements; 3) two 100-hour RPISU measurements made six months apart and 4) two-month, six-month, and twelve-month Terradex

Corp. Track Etch[®] measurements. The comparisons were made to indicate how accurately these techniques predict RPISU average annual radon-daughter concentrations in Edgemont, South Dakota.

We made five-minute air filter radon-daughter measurements in 84 buildings in Edgemont, South Dakota. Annual average radon-daughter concentrations were previously determined for these buildings from six 100-hour RPISU measurements. We also determined average radon concentrations in 50 of these buildings using Terradex Track Etch[®] detectors. The standard deviation of the difference between the natural logarithms of the RPISU annual averages and the logarithms of the air filter measurements (S.D.-ln) was 0.52. This S.D.-ln is considerably smaller than the S.D.-ln reported by ALARA (1980) at Grand Junction, Colorado, presumably because many air filter measurements in Edgemont were disregarded because of short turnover times or high wind speeds. Using the S.D.-ln of 0.52, we calculated that there would only be a 5 percent probability in Edgemont that the RPISU annual average would be greater than 0.015 working level^(a) if the five-minute measurement were equal to 0.010 WL. This indicates that the procedure used in Edgemont of clearing buildings from remedial action if the five-minute measurement were less than 0.010 WL was reasonable. There was only about a 28 percent probability that the RPISU annual average would be less than 0.015 WL if the five-minute measurement were 0.033 WL. This indicates that the protocol which requires that an engineering assessment be performed if the average of two air filter measurements is greater than 0.033 WL is also reasonable.

We have entered the Edgemont radon, radon-daughter, and average hourly meteorological data for the period from September, 1980, through May, 1982, into a computer memory data base. Initial calculations have shown no appreciable linear correlations between indoor radon-daughter concentrations and 1) wind speeds, 2) barometric pressures, 3) pressure changes, 4) outdoor temperature, and 5) outdoor radon or radon-daughter concentrations. Wind speed was the only meteorological variable that showed even the slightest indication of a correlation with indoor radon-daughter concentrations. The linear correlation coefficient squared (coefficient of determination), r^2 , between radon-daughter concentrations and wind speeds was only about 0.04. It appears that the variation in the radon-daughter concentration between buildings was so great that it overshadowed the effects of the other variables on the concentration.

There was also little linear correlation between the calculated turnover times of the radon daughters in the buildings and the wind speed. The turnover time is the residence time of the radon daughters in the building before plating out or being exchanged with outside air. The turnover time can be calculated from the disequilibrium between any two of the daughters, ^{218}Po , ^{214}Pb , and ^{214}Bi , using equations given by Morken and Scott (1966). The calculated turnover time becomes negative if the measured activity of a radon

* Trademark of the Terradex Corporation.

(a) One working level (WL) is defined as any combination of short-lived radon decay products in 1 liter of air that will result in the ultimate emission of alpha particles with a total energy of 130 billion electron volts.

daughter becomes lower than its daughter because of experimental error or variations in the plateout rate. The absolute value of the calculated negative turnover time becomes smaller for larger departures from equilibrium.

The only linear correlations found between turnover times and wind speeds were small correlations ($r^2 = 0.08$ to 0.23) between the absolute value of the negative turnover times (calculated from the ^{218}Po - ^{214}Bi pair) and the wind speeds for the same hour, the previous hour, and the second previous hour to the radon-daughter measurements. These correlations existed only for daughter measurements made when the wind speed was less than 8 mph or that showed absolute values of negative turnover times greater than 100 minutes. On the average, these negative turnover times were longer for higher wind speeds. This could be because the exchange of indoor air (having lower ^{218}Po than ^{214}Pb activities) with outdoor air (having higher ^{218}Po than ^{214}Pb activities) would tend to raise the ^{218}Po activity compared to the ^{214}Pb activity in the indoor air, thereby lengthening the negative turnover time. The air-exchange rate is expected to be more rapid at higher wind speeds. Of course, if the exchange rate became rapid enough, the influx of radon from the soil and possibly from building materials could cause the ^{218}Po activity to become greater than the ^{214}Bi activity, and the calculated turnover time would become positive and short. When we included air filter measurements made when the wind speed was greater than 8 mph, or those that showed negative turnover times shorter than 100 minutes in the data set, the correlation between the negative turnover times and the wind speeds disappeared. The above results indicate that the natural variations in the turnover times between buildings largely overshadowed those variations due to wind speed.

It appears that effects of building-to-building variations on radon-daughter concentrations and/or turnover times must be eliminated, or at least greatly reduced before the effects of changing ambient meteorological conditions can be determined. One method of doing this is to divide the measured radon-daughter concentrations by the annual average radon-daughter concentrations to obtain ratios that indicate how much the measured concentrations are above or below normal. However, the only linear correlations observed between the air-filter-to-RPISU ratios and wind speeds, pressure, and pressure changes in the 84 Edgemont buildings were small correlations with wind speed. The linear correlation coefficient squared, r^2 , between the air filter to RPISU ratios and the average wind speeds for the two hours previous to the air filter measurements was 0.09.

The above results indicate that building-to-building variations are too great to permit us to determine the effects of ambient conditions upon indoor radon-daughter concentrations from single measurements in a series of buildings. We will have to use a large number of measurements in a limited number of buildings to determine the effects of ambient conditions upon indoor radon-daughter concentrations.

Projected Work for Next Year

Radon and radon-daughter concentrations will be measured continuously for at least six months using a variety of measurement techniques in two test houses in Edgemont, South Dakota. We will measure these concentrations to determine the accuracy with which annual average indoor radon-daughter measurements can be estimated from short-term radon or radon-daughter measurements, and to determine the effect of ambient conditions during the short-term measurements upon

the accuracy of the estimated annual averages. These measurements will also be used to evaluate long-term radon and radon-daughter measurement techniques for Subtask C. Because one of the test houses will be occupied, we can study the effect of a normal range of operating conditions upon the measurement techniques. The second house will be unoccupied so that conditions can be controlled.

Radon concentrations in the houses will be measured using Kodak and Terradex Track Etch[®], grab samples in scintillation flasks, Eberline continuous radon monitors, and Passive Environmental Radon Monitors (PERM). We will measure radon-daughter concentrations using thermoluminescent dosimeter (TLD) chips, five-minute air filters (Thomas method), Alpha Nuclear Continuous Radon Daughter Monitors, and RPISU's. Outdoors, we will measure wind speed, humidity, temperature, and radon concentration. Indoors, we will measure air exchange rate (using helium as tracer), aerosol concentration, temperature, humidity and air motion.

Measurements will also be made in radon chambers to determine the effect of ambient conditions upon the accuracy of short-term radon-daughter measurement techniques.

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Subtask B. Yellowcake Emission from Ventilation Exhausts, (E. A. Lepel)

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

Accomplishments During Past Year

The available literature from the Environmental Protection Agency (EPA) and Argonne National Laboratory (ANL) have been reviewed. The following highlights should be noted: 1) particulate emissions from yellowcake drying and packaging stacks have the second highest potential for human exposure next to tailing piles; 2) there is no prescribed procedure for sampling yellowcake drying and packaging stacks, although modifications of EPA methods 1 through 5 (40 CFR 60, Appendix A) have been used; 3) the emission rate of drying and packaging stacks may vary by a factor of 2 to 5 depending on various operating parameters; 4) the aerodynamic mass median diameter of yellowcake emissions has been found to be 1.62 μm for packaging and 1.19 μm for drying exhaust stacks; 5) of the emissions observed, the respirable fraction (<2.5 μm diameter) in the packaging stack was 69%, and was 90% in the drying exhaust stacks; 6) the present use of EPA methods for isokinetic stack sampling does not necessarily measure the change in emission rate with time.

A series of recommendations were made in the literature for a general stack-testing and emission-rate calculation scheme. They were: 1) a large number of samples need to be collected from each stack over a period of a year, under each conceivable operating mode to obtain the range in emission

rates; 2) tests should be done in accordance with EPA methods; 3) for each stack, a record of operating time in each of the above conditions, needs to be maintained; 4) a total annual source term could then be determined by summing the emission rates under each operating mode; 5) the uranium daughters would be determined from a blended sample that represents the sampling year. The ideal analysis method would be a continuous monitor that is reliable, inexpensive and reasonably accurate. A continuous monitoring system has been proposed by ANL but has not been fully developed.

At present, under NUREG Guide 4.14, Rev. 1, uranium mills are required to obtain a representative sample of the effluent from the yellowcake drier and packaging stack under normal operations at least once per quarter. Discussions with a few mill operators indicate that they meet the federal requirements by taking one stack sample per quarter using EPA methods 1 through 5. Following these procedures allows the collection of isokinetic and representative samples, determination of stack gas velocity, and stack release rates for uranium, ^{230}Th , ^{226}Ra , and ^{210}Pb . The stacks are typically sampled for five to seven hours to obtain sufficient samples for analysis. Our first impression is that one sample of five to seven hours duration does not fully represent the possible changes in yellowcake emission due to changes in operational mode within the plant. Also, no estimate is available for yellowcake releases during periods when emission control equipment breaks down.

Therefore, we began laboratory evaluations to consider a real-time continuous sampling and measurement method for yellowcake, a system for taking grab samples with subsequent measurement, and an integrating sample collection method with subsequent analysis. There appear to be two distinct processes that must be considered. The first is collection of an isokinetic and representative sample and the second is accurate, economical analysis of the samples collected. It may be preferable to perform these two functions either by using a single self-contained unit for sampling and analysis in situ or by separating the two processes into in situ sampling followed by laboratory analysis. This choice will depend to a great degree on the method of analysis employed.

Projected Work for Next Year

We will complete the laboratory evaluation and subsequent determination of possible interferences and issue a topical report. Next, we will start a field verification program at an active uranium mill. We anticipate that a final report concerning this subtask will be completed late in FY 1984. This report will discuss the methods for monitoring yellowcake which were evaluated and will recommend a method or methods for adequately monitoring yellowcake releases from exhaust vents.

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

Objective

We are to determine which, if any, of the available long-term radon and radon-daughter measuring devices is the most accurate and cost-effective.

Accomplishments During Past Year

The topical report Evaluation of Experiences in Long-Term Radon and Radon Daughter Measurements, (NUREG/CR-3004, PNL-4509), was completed and sent to NRC on October 29, 1982. This report discusses side-by-side measurements of indoor radon and radon-daughter concentrations made by PNL in Edgemont, South Dakota, and compares these measurements with side-by-side measurements made by other investigators at other locations to indicate the accuracy of various radon and radon-daughter measuring devices for determining average annual concentrations of radon daughters.

We have measured average annual radon concentrations using Terradex Track Etch® Type F detectors in 50 buildings in Edgemont, South Dakota, in which the average annual concentrations of radon daughters were also measured using Radon Progeny Integrating Sampling Units (RPISU). The standard deviation of the differences between the natural logarithms of the Track Etch® radon concentrations and the logarithms of the RPISU radon-daughter concentrations ($S.D.-\ln$) was 0.37. The Track Etch® radon concentrations can be converted to radon-daughter concentrations by dividing by the average ratio between the Track Etch® and the RPISU concentrations. Using the above $S.D.-\ln$, and the Track Etch® radon-daughter concentrations calculated in this manner, we calculated that if the Track Etch® radon-daughter concentration is 0.010 WL, there should be only a 14 percent probability that the RPISU average would be greater than 0.015 WL, and only a 3 percent probability that the RPISU average would be greater than 0.020 WL. The Track Etch® and RPISU data from Edgemont, South Dakota, indicate that if year-long Track Etch® measurements had been made in all the buildings in Edgemont, and if buildings had been cleared from remedial action when the Track Etch® averages were less than 0.010 WL, then about 61 percent of the buildings would have been cleared from remedial action, and only a few of these buildings would actually have had average RPISU concentrations greater than 0.015 WL. A considerable number of buildings would have been scheduled for engineering assessment that actually had annual averages less than 0.015 WL, but the cost of each engineering assessment would have been comparable to the cost that is now being expended at Edgemont to perform each year-long RPISU measurement series.

The $S.D.-\ln$ values between the following were similar to the $S.D.-\ln$ between the Track Etch® radon measurements and the RPISU radon-daughter measurements at Edgemont: 1) the Track Etch® radon measurements and the RPISU radon-daughter measurements reported by ALARA for Grand Junction, 2) the PERM radon measurements and the MOD-225 radon-daughter measurements made by Mound Facility at Canonsburg and Middlesex, and 3) the PERM and Track Etch® radon measurements made by Mound Facility at Salt Lake City.

The fact that the $S.D.-\ln$ between the PERM and Track Etch® radon measurements was very similar to that found between radon concentrations measured by one technique and radon-daughter concentrations measured by another, suggests that radon measurements could predict radon-daughter concentrations, and that variations in the radon-daughter disequilibrium would not cause a large increase in the inaccuracy of the predicted concentrations. However, many more measurements would be needed to confirm this suggestion. These measurements also suggest that the conclusions drawn from the Edgemont data can be applied to other locations and to the PERM radon analyzer. However, a much larger $S.D.-\ln$ of 0.76 was found between PERM radon and MOD-225 radon-daughter measurements made by Mound Facility in rooms having very different ventilation

conditions in Salt Lake City. This suggests that in certain situations, radon concentrations cannot be used to predict radon-daughter concentrations. A much larger S.D.-ln of 0.84 was also found between the PERM and Track Etch* radon measurements made by Mound Facility at outdoor locations. This indicates that at least one of these techniques did not provide accurate outdoor radon measurements under at least some conditions. Many more side-by-side radon and radon-daughter measurements are needed to confirm these preliminary conclusions.

Projected Work for Next Year

We will use radon-daughter measurements in radon chambers to evaluate the effects of environmental factors upon the accuracy and operating characteristics of long-term radon-daughter measuring devices. We will measure radon and radon-daughter concentrations continually, for at least six months in the two test houses in Edgemont, South Dakota. The measurements that will be performed are listed in this report under Subtask A of Task 6. The radon-daughter concentrations measured by the various long-term measuring devices will be compared to each other and to concentrations measured on five-minute air filters to determine the accuracy of the long-term devices. We will determine the effect of environmental parameters, such as humidity, aerosol concentrations, and circulating fans upon the accuracy of the measurements of radon-daughter concentrations by these devices. The radon concentrations measured using long-term measurement techniques will be compared to the measured radon-daughter concentrations to determine whether radon concentration measurements can be used to estimate average radon-daughter concentrations, and to determine which, if any, environmental parameters can cause unacceptable errors in the estimated radon-daughter concentrations.

On the basis of the above tests, we will document the accuracy, reliability, and limitations of long-term radon-daughter measuring devices. We will also recommend procedures that should be followed when radon-daughter measurements are made in the field.

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

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

Accomplishments During the Past Year

Existing measurement capabilities for U, ^{230}Th , ^{226}Ra , and ^{210}Pb have been compared with various limits of detection suggested by the NRC in Reg. Guide 4.14 for uranium mill environmental monitoring programs. Detection limits recommended by the NRC are for air, biota, soil, and water samples.

Existing methods of wet chemical analysis can meet these detection limits and require little sample preconcentration. However, these analytical techniques are both time consuming and relatively costly, and thus, may be inappropriate to use for site evaluations where the analysis of a large number of samples is required. For these reasons, we have placed emphasis on the evaluation of instrumental procedures that require minimal sample processing, provide a short analysis time, can be utilized in mobile laboratories, and that are less costly than wet chemical methods. Our results indicate that by using planar intrinsic germanium detectors with large surface area we can achieve the best combination of selectivity and sensitivity. Present limitations of this approach in meeting suggested detection limits include 1) air samples require operation of a high volume air sampler for several days, 2) the detection limits for these radionuclides in biota cannot be reached, and 3) the detection limit for ^{230}Th in soil cannot be reached.

Projected Work for Next Year

We will modify an existing large surface area, planar, intrinsic germanium detector system to determine how much we can increase measurement sensitivities. Our modifications will involve construction of increased shielding to lower the counting system background. Preliminary calculations indicate that these changes may lower the detection limits of the isotopes of interest by a factor of 5 to 15. Such an increase in sensitivity will significantly expand the application of instrumental counting capabilities to effluent and environmental monitoring needs.

A topical report will be issued which presents the following information: 1) a comparison of existing analytical procedures for sensitivity and comparison to regulatory standards, 2) a discussion of improvements in collection and analytical sensitivity, and 3) a recommendation for field and laboratory measurement procedures.

We will defer the field verification studies to be conducted in coordination with Subtasks F and G to complete this subtask until FY 1985 due to limited funding.

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

Objective

In this subtask we are to 1) evaluate and compare well-logging techniques with core sample analytical results for the measurement of uranium and its progeny in uranium tailings, 2) determine the effects of interferences (high natural concentrations of uranium decay chain products and variations of ^{40}K concentrations) on the response of the well logging instruments, and 3) recommend an acceptable procedure which can be applied to present and future uranium mill tailings remedial action programs.

Accomplishments During the Past Year

During the past year we made progress toward a final report to suggest the best methods for estimating subsurface ^{226}Ra concentrations at remedial action sites. We conducted a thorough literature search for methods that are currently used to estimate radionuclide concentrations in boreholes. Most of

the work with boreholes has been done by the uranium exploration industry. Some of the other methods we reviewed originated from high-level waste management programs. A field trip was also made to field test methods by using an intrinsic germanium detector and a 3 in x 3 in thallium-activated sodium iodide NaI(Tl) detector to gamma log two boreholes through a tailings pile. This took place at Silver King Mine in Edgemont, South Dakota, in June, 1982. Five core samples were obtained from one hole, but core samples were unavailable from the second. We analyzed the core samples from various depths for ^{226}Ra concentrations and compared these data with the concentrations at the same depths estimated from the borehole gamma-logging data.

A topical report, Review of Well-Logging Techniques (NUREG/CR-3186, PNL-4634), was written and delivered to NRC at the end of February, 1983. This report documented several methods which could be used on remedial action programs to measure subsurface ^{226}Ra concentrations, and to meet final EPA cleanup standards. Present EPA regulations require that ^{226}Ra concentrations in soil (averaged over any 100 m surface area) resulting from residual radioactivity, shall not exceed 5 pCi/g (plus background) averaged over the 15 cm surface layer and 15 pCi/g (plus background) for each subsequent 15 cm layer below the 15 cm surface layer. The ^{226}Ra concentration in a layer can be determined either by the gamma-ray analysis of soil from core samples, from drill cuttings, or by borehole logging. However, 30 years of borehole logging by the uranium industry have demonstrated that the location and characterization of uranium ore deposits by borehole logging is cheaper, less labor intensive and more precise than sample analysis. This is especially true when core retrieval is difficult and only incomplete cores are recovered.

The uranium industry has primarily used NaI(Tl) detectors for borehole logging, but has also occasionally used intrinsic germanium detectors. Their experience indicates that these two types of detectors should be used in remedial action programs. Sodium iodide (Tl) detectors have generally been used because they were the first commercially available scintillation-type detector. However, the resolution of NaI(Tl) detectors is not good enough to measure uranium daughters other than ^{214}Bi and ^{214}Pb , so they cannot be used to distinguish between tailings material and ore. Therefore, if a NaI(Tl) detector measures ^{226}Ra concentrations greater than the regulations permit, a soil sample must be analyzed using either a germanium diode or chemical methods to determine whether it is in fact tailings material.

During the calibration of a borehole detector, the environment to which the detector will be exposed in the field must be duplicated in order to obtain valid efficiency factors. The dead time of a NaI(Tl) detector system must also be considered during calibration and borehole logging. If the dead time approaches 50%, significant errors may remain even after mathematical corrections have been made. Germanium detector systems should not exhibit significant dead times due to radionuclide concentrations encountered during remedial action programs.

At present, there are seven Department of Energy (DOE) calibration facilities present in the United States for the calibration of downhole detectors. The facilities are supported by DOE for the uranium exploration industry and are considered applicable to the calibration of downhole detectors for use in remedial action programs.

A careful review of borehole logging techniques indicates that many of the tools and methods used by the uranium exploration industry for borehole logging can be applied to remedial action projects. Many of the problems that can be expected to be encountered on remedial action programs have already been encountered by the uranium exploration industry. In order to save time and effort, suitable solutions that were applied to these problems by the uranium mining exploration industry should also be applied to remedial action programs. However, several problems must be considered in remedial action programs, which are not always considered in uranium exploration. Some of these are 1) measurements are made near the surface and at the base of the borehole, 2) auger drills that are commonly used can cause smearing of the tailings layer, and 3) radon is released from the borehole causing possible disequilibrium between radium and its radioactive decay daughters.

The calculation of ^{226}Ra concentrations from borehole logging data is further complicated by the fact that even when using a collimated detector, some of the gamma-rays measured originate from depths that are different from that of the detector. The smearing of the signal that results is called geologic impulse response (GIR) or radius of investigation. Various methods have been developed to minimize the effects of GIR, and these have been used extensively by the uranium exploration industry. Two methods of particular interest (Gamlog and Exact Inverse Filtering) were discussed in the topical report.

Projected Work for Next Year

Two field trips are planned, one to Edgemont, South Dakota, and the second to the DOE calibration facility at Grand Junction, Colorado. The purpose of the Edgemont trip is to collect data using a down-hole intrinsic germanium detector around dwellings where subsurface ^{226}Ra concentrations at or near background levels are known as a function of depth. This will determine the suitability of the intrinsic germanium detector for low level work. We will also obtain data using NaI(Tl) detectors and grab sampling techniques and compare these data to the results calculated from intrinsic germanium data. The purpose of the Grand Junction trip is to use an intrinsic germanium detector and possibly an NaI(Tl) detector to log the calibration wells at this facility and compare the resultant ^{226}Ra concentration estimates with the well-documented calibration sources.

Completion of the final report is scheduled for January 31, 1984. This report will recommend procedures to accurately estimate subsurface ^{226}Ra concentrations at remedial action sites.

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

Objective

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

Accomplishments During Past Year

A data base which is expected to be typical of those collected near tailings piles was selected for analysis by geostatistical methods. The data chosen for the study are radium (^{226}Ra) concentrations in soil of the Cottonwood area of Edgemont, South Dakota. This data base has a number of characteristics that make it useful as a tool for evaluating scenarios for spatially distributed phenomena. Samples were collected at a relatively large number of sampling points. Some sections of the sampled area are more intensively sampled than other sections. The area is adjacent to old mill tailings piles, and therefore, we expect that there is a spatial distribution of radium concentrations caused by windblown materials. Background data are available in the form of soil cores collected from depths where disturbance by human activities is unlikely. Finally, the data were recently collected and the persons who collected them are readily available to answer any questions.

These samples have been analyzed using Battelle computer programs for Kriging, which is a geostatistical technique for determining spatial inventory and distribution of any material. We found that in this particular data set, the major source of statistical variability was the replication error at any given sampling point, and therefore, the amount of sampling and the sampling scheme appeared to have little effect on the estimates of spatial distribution. For situations like Edgemont, the accuracy of the spatial pattern derived by Kriging is only slightly affected by the number of samples used, and thus a small number of samples taken on a widely spaced grid is the most efficient sampling plan. While accuracy of the spatial pattern will not be greatly enhanced by using many samples, it is possible that a large grid sampling plan will not give sufficient detail for planning cleanup activities. Investigation into this possibility is continuing.

Projected Work for Next Year

A topical report Recommended Sampling Strategies for Spatial Evaluation of Windblown Contamination Around Uranium Tailings Piles will be issued in July, 1983.

Due to limited funding, continuation of this subtask will be deferred until FY 1985.

Projected Work for FY 1985

The recommended procedures will be used to guide PNL surveys of windblown tailings at at least one inactive tailings site scheduled for UMTRA. This would hopefully be conducted concurrently with surveys being performed by the remedial action contractor for the UMTRAP, and be conducted in coordination with field verification studies conducted under Subtasks D and G. In addition, we have proposed to further evaluate data from multiple surveys of selected uranium mill tailings sites and select one or possibly two sites to revisit. We will take field measurements to "fill in the holes" in existing data and allow comparisons between sequential surveys. This will enable us to evaluate sampling strategies over time.

Subtask G. Sampling Techniques for Detecting Contamination of Surface and Ground water by Tailings Leachate, (W. C. Weimer, R. R. Kinnison)

Objective

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

Accomplishments During the Past Year

We completed the evaluation of existing analytical methods for measuring selected stable elements and radioactive uranium-daughter products. Existing methods, which are widely available, are satisfactory for meeting the anticipated monitoring needs for detecting contamination of both surface and ground water. We identified a void in the existing base of data for ground water monitoring programs related to the statistical design of monitoring programs. Therefore, an existing large ground water data base gathered as part of the Hanford ground water monitoring program has been selected to use for the development and statistical evaluation of ground water sampling protocol.

Projected Work for Next Year

The Hanford ground water data will be used to test a number of statistically derived sampling strategies that evaluate well placement, number of wells, and sampling times. We will use the Kriging geostatistical technique to evaluate the number of sampling points that are required with respect to the desired qualities of the radionuclide inventory estimate and the radionuclide concentration-distribution estimate.

A topical report will be issued which presents the following information: 1) an assessment of available analytical methodologies for measuring selected stable elements and radioactive uranium-daughter products in natural waters, 2) a protocol for monitoring the surface and ground waters near uranium recovery sites, and 3) a statistical evaluation of the development of this protocol.

Field verification studies to be conducted in coordination with Subtasks D and F are deferred until FY 1985 due to limited funding.

Subtask I. Radon Flux Comparisons with ^{226}Ra Concentrations in Surface and Subsurface Soils, (J. A. Young)

Objective

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

Accomplishments During Past Year

Plans and preparations have been made for a field trip to Edgemont, South Dakota, in support of this recently initiated subtask. We will measure radon fluxes from locations where ^{226}Ra concentrations in surface and subsurface soil have been measured previously.

Projected Work for Next Year

Radon fluxes will be measured at locations in Edgemont, South Dakota, where ^{226}Ra concentrations have been logged in boreholes and determined from the analysis of core samples. We will also measure radon fluxes at locations in Edgemont where comparisons are to be made between ^{226}Ra concentrations measured in core samples and those measured using borehole logging techniques as part of Subtask E.

TASK 7. TECHNICAL PROGRAM MANAGEMENT

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

FIN B-2370 (Task 7)

OBJECTIVE

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

ACCOMPLISHMENTS DURING THE REPORTING PERIOD

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

- Arranging and participating in an NRC Headquarters review of the Long-Term and Interim Stabilization Tasks (B-2370, Tasks 1 and 2, 21 April).
- Arranging and participating in PNL presentations at the workshop on hydrological and geochemical modeling held in Silver Spring, Maryland on 14 and 15 June.
- Arranging and participating in an NRC Headquarters Review of Tailings Dewatering, (B-2370, Task 3); Tailings Neutralization (B-2370, Task 4); Disposal Techniques Allowing Contact with Ground Water (B-2370, Task 5); and Assessment of Leachment Movement From Uranium Mill Tailings (B-2292).
- Assisting in writing and reviewing revisions to FY-82 189 submissions on B-2370, Stabilization, Engineering, and Monitoring Alternatives Assessment for Improving Regulation of Uranium Recovery Operations and Waste Management; B-2269, Attenuation of Radon Emissions From Uranium Tailings Piles; and B-2292, Assessment of Leachate Movement From Uranium Mill Tailings.

- Submitting the foregoing three 189s for FY-83 in June and July plus B-2379, Methods for Minimizing Ground Water Contamination in In Situ Leach Uranium Mining. A new 189 was also submitted, entitled "Assessment of Hydrologic Transport with Chemical Interaction for Uranium Recovery Research."
- Developing and adopting a program-wide Quality Assurance Plan.
- Developing and adopting a program-wide Quality Assurance Procedure for checking calculations.
- Developing and implementing a computerized program-management system to assist in the timely completion of reports to the NRC, and using it to help meet schedule with a high percentage of our deliverables.
- Assisting in preparing and submitting the final revision to the FY-83 189 for B-2370.
- Arranging and participating in an NRC review of the uranium recovery research program at PNL.
- Reviewing the drafts and final documents of the reports produced by the uranium recovery program (listed elsewhere in this report).
- Preparing and submitting 12 monthly administrative reports and four quarterly technical progress reports.

With the retirement of L. C. Schwendiman at the end of August 1982, management of the Uranium Recovery program was assumed by Dr. G. W. Gee during a transition period in September. Dr. M. G. Foley became program manager on 1 October 1982. In the same period, several task leader assignments changed, reflecting changes in staff or changes to align staff expertise more closely with the evolving technical direction of a task. The revised task leader assignments have already been shown in Figure 1.

PROJECTED WORK FOR THE NEXT YEAR

In addition to managing the technical and budgetary schedules, and submission of periodic administrative and technical progress reports, the program manager will assure that:

- Revised 189's including preliminary plans for FY-85 work, will be prepared and submitted.
- The status of uranium recovery waste management technology will be addressed, and short-term and long-term research needs determined so that the PNL program properly meets the needs of NRC-RES.
- Reviews of PNL work be internal and external peer reviewers and by NRC will be arranged and conducted.

ATTENUATION OF RADON EMISSION FROM URANIUM MILL TAILINGS

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

FIN B-2269

OBJECTIVE

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

ACCOMPLISHMENTS DURING REPORT PERIOD

Task 3. Measure Radon Diffusion Coefficients in Candidate Cover Soils

Objective

In this task we are to measure the diffusion coefficients of radon in candidate soils for covering uranium mill tailings and to estimate the range of values that can be expected at tailings sites.

Accomplishments During Report Period

We measured diffusion coefficients for radon in 34 soils that are candidates for covering uranium-mill tailings piles to reduce radon emission. Measured values were extrapolated to those expected under the long-term conditions for soil-moisture contents at tailing sites. Values for silty sands and clayey sands ranged from 0.004 to 0.06 cm²/s, whereas values for inorganic silts and clays ranged from 0.001 to 0.02 cm²/s. A camera-ready copy of the report, Radon Diffusion in Candidate Soils for Covering Uranium Mill Tailings, NUREG/CR-2924, authored by W. B. Silker and D. R. Kalkwarf was sent to NRC.

Task 4. Field Test Radon Attenuation by Cover Soils

Objective

In this task we compare radon diffusion coefficients evaluated for 0.1 m depths of soils under laboratory conditions, with diffusion coefficients

evaluated for several-fold greater depths of the same soils used to cover uranium mill tailings.

Accomplishments During Past Year

Measurements of radon flux from covered and uncovered tailings at the Dawn Mining Co., Ford, Washington, were used to evaluate radon diffusion coefficients in the cover soils. The results were compared with measurements of diffusion coefficients for radon in 0.1-m deep columns of the same soils in the laboratory. The comparison showed that radon diffusion coefficients evaluated from field data were statistically indistinguishable at the 95 percent confidence level, from those measured in the laboratory; however, the low precision of the field data prevented a sensitive validation of the laboratory measurements. A camera-ready copy of the report, Comparison of Field-Measured Radon Diffusion Coefficients with Laboratory-Measured Coefficients, authored by E. A. Lepel, V. W. Thomas, W. B. Silker and D. R. Kalkwarf, NUREG/CR-2769, was sent to NRC.

Task 5. Assess the Influence of Defects on Radon Diffusion in Cover Soils

Objective

In this task we are to 1) determine the size, shape, and frequency of defects in a soil cover which can significantly alter the radon-attenuating ability of the cover, 2) test the validity of radon-diffusion equations to predict the flux of radon through defective soil covers, and 3) identify soils to be avoided as cover material because of their proclivity to form cracks.

Accomplishments During Past Year

We conducted a literature search for methods to estimate the influence of soil-cover defects on the radon flux from covered uranium-mill tailings and for methods to avoid defect formation. These defects include shrinkage and erosion cracks, erosion piping, animal burrows, and vegetation roots. Equations based on gaseous-diffusion theory have indicated that the average radon flux from covered uranium-mill tailings can be greatly increased by certain types of cracks in the cover. If the crack penetrated the entire cover and its cross-sectional area was 1 to 4 percent of the covered area, a 10- to 30-fold increase in flux was predicted (Mayer and Zimmerman 1981; Landman 1982). If other transport mechanisms for radon also occur in the cracks, still larger flux increases were predicted (Landman and Cohen 1983).

The literature survey also revealed several techniques for reducing the probability of defect formation. Avoiding the use of montmorillonitic clay in cover soil should decrease the chance of forming shrinkage cracks (Hillel 1980), and avoiding sodic soils should avert the formation of soil piping (Gee et al. 1978). Sustained release of herbicides has been reported and recommended as a means for reducing root growth into soil covers for uranium mill tailings (Cline et al. 1982). Similarly, sustained release of rodenticides has been

proposed for controlling the formation of animal burrows in soil (Scher 1977). Layers of small stones on top of soil covering tailings has also been shown to prevent formation of animal burrows (Cline et al. 1982).

In the laboratory, we conducted experiments on model defects in soil columns to search for conditions leading to the large increases in radon flux predicted in the literature. Soil columns, 10 cm in height and 14 cm in diameter, were used as covers for reservoirs filled with radon gas from radium salts. Due to the short height of the soil columns, large increases in radon flux resulting from cracks have not been observed directly because even defect-free columns did not attenuate the flux by more than 10 percent. However, the concentration gradient of radon across the column was a more sensitive, albeit indirect, indicator of crack influence. If a 1.5 cm wide rectangular crack bisected the soil cover and penetrated the entire column, the cover was completely ineffective in reducing radon flux. The radon flux at the top of the soil column was the same as that at the bottom, and the concentration of radon in the soil gas at the bottom of the column was the same as that at the bottom of an equal-length column of air. With cracks of narrower width, larger concentration gradients of radon were established in the air-filled cracks; but these gradients were gradually reduced over periods of days, a phenomenon attributed to turbulent diffusion of radon from the crack. Even a 0.3-cm wide crack reduced the radon concentration gradient across a soil column, and hence, increased the average diffusion coefficient for radon in the column. These data are now being analyzed to estimate the potential influence of cracks in the covers for tailings.

Task 6. Devise and Test Alternative Measurements of Radon Diffusion Coefficients (RAE)

Objective

In this task, we are to devise and test an alternative to the steady-state methods for measuring diffusion coefficients of radon in soil.

Accomplishments During Past Year

We devised a transient-diffusion method to measure diffusion coefficients of radon in soils, and the coefficients obtained by this method were within 10 percent of those obtained by a steady-state method for soils with moisture saturation less than 0.6. With soils at higher moisture saturation, the two methods gave coefficients which differed by up to a factor of 2. A camera-ready copy of the report, Comparison of Radon Diffusion Coefficients Measured by Transient-Diffusion and Steady-State Laboratory Methods, NUREG/CR-2765, authored by K. K. Nielson and V. C. Rogers, was sent to NRC to describe this research.

Task 7. Predict Radon Diffusion Coefficients from Soil Properties (RAE)

Objective

In this task, we are to devise a mathematical model for predicting the diffusion coefficients of radon in soil columns based on the moisture content and pore-size distribution of the soil column.

Accomplishments During Past Year

We devised a mathematical model which predicted experimentally measured radon diffusion coefficients in soil to within a factor of 6, using only the moisture content and pore-size distribution in the soil as parameters. A camera-ready copy of the report, A Mathematical Model for Radon Diffusion in Earthen Materials, NUREG/CR-2765, authored by K. K. Nielson and V. C. Rogers was sent to NRC to describe this research.

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

Objective

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

Accomplishments During Past Year

An updated version of a A Handbook for the Determination of Radon Attenuation Through Cover Soils, NUREG/CR-2340, is being prepared. A literature survey of current and recent work in radon research was conducted and the chapter on methods for measuring radon diffusion was revised. Also, additional values of radon diffusion coefficients were incorporated into the present data base for predicting these coefficients from other soil parameters.

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

Objective

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

Accomplishments During Past Year

Data taken at the UMTRAP site at Grand Junction, Colorado, were collected in preparation for comparing field measurements of radon flux with values calculated by handbook methods.

Task 11. Describe Advective Radon Transport Through Cover Soils (RAE)

Objective

We are to evaluate the advective transport of radon with water vapor in soils and to assess the advection of radon in dry air resulting from changes in temperature and atmospheric pressure.

Accomplishments During Past Year

From theoretical calculations and a literature review, we predicted that advective radon flux from soil surfaces will occur due to gradients in air pressure, temperature and moisture in soils. The combined effect of these factors in the short term was examined by field measurements of radon flux at the Vitro Corporation uranium-mill tailings site near Salt Lake City, Utah. The results showed that radon flux varied diurnally with a maximum during the night hours (22:00 to 06:00) that was 10 percent higher than that during the day. In order to assess the long-term effects of advection, the contributing processes were examined individually in laboratory experiments. An apparatus was constructed which allowed evaluation of radon diffusion coefficients on soils in which advective air flow, temperature, and moisture content were carefully regulated. The dependence of these coefficients on advective air flow was found to follow theoretical calculations up to the maximum coefficient expected in soil. The dependence of radon diffusion coefficients on advective flow of water vapor is also being evaluated, but has required measurements on soil columns at different temperatures to obtain a range of water-vapor flow rates. This, in turn necessitated that of the diffusion coefficients be corrected to account for the effect of temperature.

In a separate dry-advection experiment, we are examining the effects of cyclic advection events. This experiment also uses experimental conditions similar to those in the above experiment; but in this case the air flows are applied to the bottom of the 60-cm tailings source so that any advective movement includes the entire tailings-cover system. Using a selected pair of critical orifices to control flow rates, air is alternately forced into the tailings column and then withdrawn from the column using compressed air and a vacuum system as the driving forces. Velocities through the experiment column are on the order of 10^{-5} cm/s, and the time for each air input or withdrawal is normally 24 hours. A buffer chamber of radon gas is used to condition all exchanged air from the column so that clean air does not dilute the input air in the tailings source region. Initial radon fluxes were widely separated (about a factor of 100), but after a number of cycles the fluxes are approaching a repeating pattern. This experiment is still in progress, and we will use it as part of an analysis of the importance of cyclic events, such as barometric pressure changes, on the effectiveness of tailings covers in radon containment.

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

Objective

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

Accomplishments During Past Year

An initial outline of the guide was prepared, and is being reviewed internally.

PROJECTED WORK FOR NEXT YEAR

This project is scheduled for completion in FY-83. During the remaining time we will complete the following activities. Results from 1) investigation of the advective transport of radon in soil covers, and 2) study of the influence of cover defects on attenuation of radon emission, will be described in topical reports on these subjects.

Projected Work for Next Year

We will make additional laboratory measurements of diffusion coefficients for radon in the soils used at the Grand Junction site. These will be used to determine diffusion-coefficient values for the exact conditions of soil porosity and moisture content used in the field. Also, additional radon-flux measurements will be made at one of the covered test plots at the Grand Junction site immediately before excavating a hole in the plot to verify long-term moisture content, compaction and radon emanation coefficient. A topical report describing this study will be prepared for NRC.

The document, Handbook for Determination of Radon Attenuation Through Cover Soils, NUREG/CR-2340, will be revised based on the results of research carried out since its preparation. We will test the validity of the methods recommended in this handbook by using field data and samples from the UMTRAP site at Grand Junction, Colorado. The results of this study will be described in another topical report. Finally, we will prepare a document in the format of an NRC Regulatory Guide which specifies acceptable methods for covering uranium-mill tailings with soil to reduce radon flux to within prescribed limits.

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ASSESSMENT OF LEACHATE MOVEMENT FROM URANIUM MILL TAILINGS

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FIN B-2292

OBJECTIVE

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

ACCOMPLISHMENTS DURING REPORT PERIOD

Task 1. Project Management

On June 16, 1982, a review of this project was conducted in Silver Spring as part of a comprehensive review of the following NRC projects: Assessment of Leachate Movement From Uranium Mill Tailings and Tasks 3, 4, and 5 of Stabilization, Engineering and Monitoring Alternatives Assessment for Improving Regulation of Uranium Recovery Operations and Waste Management. The review involved PNL staff associated with the above mentioned tasks and responsible NRC personnel. The questions and discussion indicated no particular problems with the direction or emphasis of the program.

On June 15, we presented an overview of hydrological and geochemical modeling related to uranium recovery work. In addition to discussing hydrological and geochemical models applicable to uranium mill tailings, we outlined the methodology for expanding the thermodynamic data base that is pertinent to these operations.

R. L. Erikson gave a presentation at the Fifth Symposium on Uranium Mill Tailings Management held at Fort Collins, Colorado. This presentation concerned a mineralogical investigation of the interactions of acid leachate with soil minerals at the Lucky Mc Pathfinder Gas-Hills Mill in Wyoming. In addition, a paper addressing the same subject was published in the symposium proceedings (Erikson and Sherwood 1982). S. R. Peterson prepared and gave a presentation at the National Soil Science Society of America meetings in Anaheim, California (Peterson and Serne 1982). This presentation dealt with the interactions between sediments and uranium mill tailings and compared geochemical modeling predictions in this area to laboratory and field data.

The report on the User's Manual for DIGRID was completed, reviewed, and published. The topical report on The Long-Term Stability of Earthen Materials in Contact With Acidic Tailings Solutions was also published by the NRC.

In addition, our laboratory progress report entitled Laboratory Measurements of Contaminant Attenuation of Uranium Mill Tailings Leachates was finished. The 189 for FY-83 was written and a manpower/cost outline was prepared based upon the funds received for FY-83. We began writing the topical report for June which deals with geochemical modeling as an aid to understanding contaminant migration.

An abstract was prepared for presentation at the National Agronomy meetings. S. R. Peterson and R. J. Serne will present "Predictive Geochemical Modeling of Permeability Column Effluents" at the annual ASA-CSSA-SSSA meetings to be held August 14-19 in Washington, D.C.

Task 2. Laboratory Analysis and Experiments

We conducted experiments to determine changes in liner materials during contact with acidic tailings solutions. Our evaluation of the mineralogical stability of liner materials involved three studies: 1) batch-type experiments, 2) column-type experiments, and 3) a field study. The stability and the potential mineralogical alteration of liner materials in contact with acidic tailings solutions were determined by comparing the mineralogy before and after contact with tailings solutions. A major conclusion drawn from the three mineralogy studies is that the results of the laboratory experiments and field data are similar and complement the permeability studies, laboratory chemistry studies and geochemical modeling predictions.

Liner failure (i.e., an increase in the permeability of the liner material) was not found to be a problem when various acidic tailings solutions leached through liner materials for periods up to three years. Liner materials containing over 30 percent clay showed a decrease in permeability with time in laboratory columns. The materials with high clay content that we tested appear suitable for lining tailings impoundment ponds. One clay liner composed of a silty clay loam, decreased in permeability from 1/2 to over two orders of magnitude depending upon the given column and the contacting solution. Some soil materials that we tested contained less clay (approximately 7 percent) and showed decreases in permeability. Only a sandy loam with approximately 9 percent clay and little carbonate (approximately 0.2 percent), exhibited an increase in permeability over the time span of the study. The decreases in permeability were attributed to pore plugging resulting from the precipitation of minerals and solids and to soil particle dispersion (Peterson et al. 1982).

The clay liner and the overburden from Morton Ranch, Wyoming, exhibited a residual long-term buffering capacity that maintained the pH values of the column effluent at a higher level than the influent pH values. Because of the residual buffering capacity, the pH of the tailings solution migrating away from the tailings impoundment area can have a higher pH than the original tailings solution, even after the initial buffering capacity of the soil has been exhausted. We need further studies to explain the chemical reactions that

account for the residual buffering capacity of soils and liners; however, a likely cause is the redissolution of Fe and Al hydrous oxides. Redissolution of Fe and Al hydrous oxides consumes hydrogen ions.

We also began the selection of analytical methods for determining uranium mill tailings chemistry. In general, inductively coupled plasma (ICP) is suggested for macro-cation analysis, while graphite furnace atomic absorption (AA) is recommended for many trace metals. We have demonstrated that with the AA we can meet MPC or lower concentrations for As, Cd, Cr, Cu, Fe, Mo, Pb, Mn and V. Selenium (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. Table 2 lists the preferred analytical techniques for selected constituents of uranium mill tailings.

Because high-acid, high-salt tailings solutions present significant analytical problems for metals analysis, we directed our preliminary efforts towards determination of recommended techniques and practical detection limits. For metals analysis, we determined that dilution of the tailings and column effluent solutions was required to reduce spectral, physical, and non-specific absorption interferences to acceptable ranges to allow accurate results. The dilution procedure directly increased detection limits for most trace metals to the range 10-50 parts per billion (ppb). Thus, for routine analysis of large numbers of samples, the practical detection limits for most trace metals are at or slightly exceed the recommended maximum concentration levels for drinking water. In general, the trace metals concentrations of the acidic U mill tailings solutions are present in the low parts-per-million range and thus readily measured. Notable exceptions include Ba and Pb which are present at <50 parts per billion. In general, the neutralized effluents emanating from the sediment columns show trace metal concentrations at or below the practical detection limits (10-50 ppb). Notable exceptions in the initial effluents include Se and Mo and perhaps Zn.

Ion chromatography can detect sulfate, chloride, fluoride, nitrate, and bromide down to the low parts-per-million (ppm) level in the acid tailings solution and sediment-contacted effluents. Sulfate predominates (2,000 to 20,000 ppm) in the solutions and chloride is present at 100-300 ppm concentrations. Nitrate varies from the low ppm's to 500 ppm, fluoride from less than detectable to 10 ppm and Br is usually not detectable (<0.5 ppm).

Amorphous stripping techniques were selected to help elucidate the types of compounds, within the sediments and clay liners, that bind to the trace metals and macro-constituents. We selected procedures that separate the phases outlined below, because they are important to the geochemistry of contaminant migration from uranium disposal sites. Peterson and Krupka (1981) identified many of these phases as possible precipitates when tailings liquors contact the surrounding geologic media; in addition, precipitates can act as scavengers of trace metals (Jenne 1968). We reviewed the existing literature to determine the techniques with the greatest selectivity. The phase differentiations and their recommended extraction techniques are listed below.

TABLE 2. Preferred Analytical Techniques for Selected Constituents of Acid Uranium Mill Tailings Related Solutions

Constituent	Method ^(a)	Detection Limit (mg/l)	Comments
Li	ICP	0.01	Typically found at 0.1 to 1 mg/l; no significant spectral interfaces observed
B	ICP	0.05	Typically found at 0.1 to 0.5 mg/l Fe; U spectral interferences
HCO ₃ -CO ₃	Titration	NM ^(b)	Only found in effluents from sediment contact where acidity has been neutralized
NO ₃	IC	1	Typically found at 1 to 300 mg/l
F	IC	0.5	Typically found at <0.5 to 20 mg/l
Na	ICP	1	Typically found at 50 to 500 mg/l; precision of ICP is poor ±20 to 30%; Fe and U spectral interferences; AA flame more precise
Mg	ICP	3.0	Typically found at 50 to 2000 mg/l; U spectral interference is large
Al	ICP	0.05	Typically found at <0.05 to 500 mg/l; large U and Fe interferences; AA/GF may be useful at low concentration
Si	ICP	0.05	Typically found at 10 to 300 mg/l; U and Fe interferences
P	IC	1.	Typically <1; should try sensitive colorimetric technique if low quantities need to be determined
SO ₄	IC	NM	Typically present at 1000 to 20,000 mg/l
Cl	IC	NM	Typically present at 50 to 500 mg/l
K	ICP	3	Typically present at 10 to 200 mg/l; Fe interferes; Flame AA more precise
Ca	ICP	0.2	Typically present at 500 to 2000 mg/l; U and Fe spectral interferences
V	AA/GF	0.05	Typically present at <0.05 to 5 mg/l
Cr	AA/GF	0.03	Typically present at <0.03 to 3 mg/l; ICP suffers from spectral interference
Mn ^(c)	ICP	0.05	Typically present at <0.05 to 50 mg/l; AA/GF as good or better
Fe	AA/GF	0.02	Typically present at <0.02 to 1000 mg/l; ICP good if Fe > 0.1 mg/l
Co ^(c)	ICP	0.03	Typically present at <0.03 to 1 mg/l; AA/GF probably better but was not checked
Ni ^(c)	ICP	0.06	Typically present at <0.06 to 2 mg/l; AA/GF probably better but was not checked
Cu	AA/GF	0.05	Typically present at <0.05 to 1 mg/l; ICP OK at Cu > 0.2 mg/l; U spectral interference
Zn ^(c)	ICP	0.10	Typically present at <0.1 to 5 mg/l; AA/GF may be better but was not checked.
As	AA/GF	0.05	Typically present at <0.05 to 20 mg/l; ICP suffers numerous spectral interferences, use Ni salt matrix modification for AA/GF
Se	AA/GF	0.02	Typically present at <0.02 to 5 mg/l; use Ni salt matrix modification for AA/GF
Sr	ICP	0.04	Typically present at 5 to 30 mg/l
Mo	AA/GF	0.06	Typically present at <0.06 to 1 mg/l; ICP OK at Mo > 0.3 mg/l
Cd	AA/GF	0.01	Typically present at <0.01 to 3 mg/l; ICP OK at Cd > 0.2 mg/l
Sb ^(c)	ICP	1	Typically present at <1 mg/l; AA/GF may be better but was not checked
Ba	ICP	0.05	Typically present at <0.05 to 0.2 mg/l
La	ICP	0.2	Typically present at 1 to 10 mg/l; U spectral interference
Pb	AA/GF	0.02	Typically present at <0.02 to 0.1 mg/l; use platform graphite furnace; ICP spectral interferences are severe.

(a) Methods

ICP Inductively Coupled Argon Plasma Emission Spectroscopy

AA/GF Atomic Absorption with Graphite Furnace

IC Ion Chromatography

(b) NM = not measured

(c) ICP is used because it is cost effective but AA/GF would probably be more accurate and sensitive. Because element is not a priority pollutant it is not necessary to spend extra money and time for more accurate analysis.

Phase	Technique
1. Exchangeable phase, carbonates, sulfates	2.5 percent acetic acid
2. Manganese hydroxides	0.1 M $\text{NH}_2\text{OH} \cdot \text{HCl}$ at pH 2 Oxalate Solution
3. Amorphous Al and Fe	Ascorbic acid and oxalate
4. Crystalline Fe	Li metaborate fusion, analysis by ICP
5. Residue	
6. Total (sample not extracted)	Li metaborate fusion, analysis by ICP

The first suite of laboratory vacuum-extractor column experiments were finished. This involved dissecting twenty-four columns that had been packed with the Morton Ranch clay liner and leached with the Highland Mill tailings solution spiked with ^{110}Ag , ^{51}Cr , ^{109}Cd , ^{210}Pb , ^{73}As , ^{75}Se , ^{54}Mn , ^{57}Fe , and ^{228}Ra . The effluent solutions from these columns were analyzed by ICP, IC, and radioanalytical techniques. We radiocounted the subsections of the dissected columns to develop a profile of where each element was adsorbed and also to ascertain those column subsections where it would be advantageous to use the stripping techniques.

Stripping techniques (selective extractions) were completed on selected subsections of the dissected columns. The stripping techniques used and the phase differentiations they provide will help determine which types of minerals--indigenous and precipitated--within the sediments bind the trace metals and radionuclides.

Three different types of data (column effluent, nuclide distributions bound to sediment as a function of depth, and selective extraction) were combined to allow us a comprehensive tool to judge the relative migration rates of the contaminants, and to provide insight into the processes that bind the contaminants. The three data types emanated from experiments involving vacuum-extractor columns, permeability cells, column radiocounting techniques, and extractions with assorted chemical reagents. These provide insight on the relative binding strength that the sediments have for the contaminants, and to what geochemical phases the contaminants are associated. The results of the various laboratory experiments allow us to summarize, in the following paragraphs, a few of the observations made this quarter with regard to the particular clay liner and particular acidic uranium mill tailings solutions used.

Arsenic (As) and chromium (Cr) were present in the acid tailings solution at levels between 0.5 and 3.5 mg/l (well above the EPA drinking-water standard of 0.05 mg/l) and readily became bound to the clay liner. Both radiotracer

and total arsenic analyses support the finding that arsenic does not seem to remobilize after the neutralizing capacity of the clay liner is exhausted. Arsenic extraction data suggest that sediment-bound arsenic, like chromium and selenium (Se), is bound to amorphous and crystalline iron and perhaps aluminum oxides. We speculate that the controlling process is anion attraction/exchange of the arsenic, chromium, and selenium partially present as anions in the solution. At low pH values the hydrous oxides are protonated $(Al, Fe)-(OH)_2^{4-x}$ such that a net positive surface charge is present to which anions are attracted. A second immobilization process could be precipitation of ferric arsenates and selenites, which are both very insoluble compounds.

Relatively speaking, arsenic is bound more firmly than chromium and selenium. The residual sediments after chemical extraction with five progressively harsher reagents contained 22 percent by weight of the originally bound arsenic and <15 percent of the chromium. Although the residual sediments contained 50 percent by weight of the originally bound selenium, there is a very mobile form present. This is evidenced by high selenium concentrations in the first samples of column effluent and detectable Se radiotracer throughout the contacted clay liner even though the majority of selenium tracer is bound in the top of the column. The selenium data suggest that perhaps several species may be present, and at least one of these is mobile and apparently slow to convert to more easily attenuated forms. Computer speciation modeling shows selenate (SeO_4^{2-}) will be more common in these solutions and perhaps is the mobile form of selenium. We suggest that the data show that arsenic and chromium do not readily remobilize after the acid tailings exhaust the clay liner's neutralizing capacity, and that a relatively mobile and somewhat pH independent selenium species exists.

Total molybdenum results show that high concentrations of molybdenum effluent occur in the first solution emanating from the columns, but little molybdenum occurs in subsequent effluents. More work is needed to identify the forms and processes by which molybdenum and selenium migrate, and we expect the next document in this series on geochemical modeling to provide additional insight. Studies of neutralization of acidic tailings solutions with various reagents such as calcium carbonate (limestone), hydrated lime $[Ca(OH)_2]$, sodium hydroxide, and sodium carbonate, also show that molybdenum and selenium are not readily removed from solution and warrant further attention (see Sherwood and Serne 1982).

Silver (Ag) is present in the acidic tailings solution at less than detectable concentrations (<0.05 mg/l), but by using the radiotracer, ^{110}Ag , we found that silver is rapidly retained in the upper 10 percent of the clay liner when acidic solution is percolated through the liner. The radiosilver that is bound in the sediments appears to be either bound strongly (on a weight basis 50 percent of the bound silver is not extractable) or associated with cation exchange sites. It appears that the silver migrates very slowly through the clay liner even after the neutralization capacity has been exhausted. Detectable silver (tracer or stable element) was not observed in any column effluent solutions.

Although few data are available on vanadium, it does not appear to be very mobile. We saw no measurable vanadium in column effluents, and the total vanadium concentration bound in the column after solution percolation shows elevated concentrations in only the top half. Although we have no selective extraction data, we speculate by chemical analogy that anionic vanadium (VO_4^{3-}) may be bound in a similar fashion as arsenic and chromium, or that the low solubility uranium-vanadium mineral, tyuyamunite, may be controlling the solution concentration of vanadium. Again, the upcoming modeling report should provide insight.

The data available from our research on transition metals such as cadmium, cobalt, nickel, zinc and copper allow us to conclude that transition metals are immobilized by the sediment during the neutralization process and remobilized as the acid front travels through the column after the sediment's buffering capacity is exhausted. The bound transition metals are associated with amorphous and crystalline hydrous oxides, especially under neutral pH conditions, and partly with cation exchange sites [especially under more acidic pH conditions, pH 3-4.5]. All of these trace metals show very low concentrations in the column effluent while the pH is near neutral and relatively high concentrations as the pH front (pH ~4) arrives at the column effluent. At this point, concentrations of these metals typically exceed the acid-tailings solution concentration, suggesting that redissolution of previously bound metals is occurring. The redissolution process of the pH front is thus similar to a snow plow that collects large quantities of snow in front of its blade. The data further suggest that a quantitative description of the chemical interactions will require expressions not only for the pH-dependent immobilization of hydrous oxides (perhaps via thermodynamic solubility relationships), but also for retardation by retention on ion exchange sites. Uranium data do not show the snow plow effect, but are pH sensitive in that increased uranium values occur in solution once the pH front breaks through. Furthermore, the distribution of the total uranium bound in the sediment at the end of the experiment shows a peak concentration just on the acidic side of the pH front. A similar uranium peak was observed in the field beneath a tailings-solution evaporation pond (Peterson et al. 1982; Erikson and Sherwood 1982).

Data on lead (Pb) show that it is retained by sediments more strongly than the other transition metals and associates much less with cation exchange sites. No detectable total lead nor ^{210}Pb was found in column effluents. Again chemical extraction results show that much of the bound lead is associated with hydrous oxides of iron and aluminum. Although data were not obtained on thorium (^{230}Th), we speculate that it will behave very similarly to lead and not migrate very far into the sediment. Sherwood and Serne (1982) found thorium was readily removed by all the neutralizing agents used.

Data on manganese, iron, and aluminum in column effluents support the importance of hydrous oxide formation when acid tailings solutions are neutralized, as a major process in lowering their solution concentration. Based on selective extraction results, cation exchange may play an even greater role than pH neutralization in lowering manganese concentrations. Cation exchange, especially in the more acidic top portions of the columns, is also important for aluminum and iron. Maximum manganese concentrations in the sediment occur

just at the leading edge of the pH-front breakthrough. Again, these data point out the need to combine the precipitation/dissolution of pH-sensitive hydrous oxides with cation-exchange relationships to best describe mass transport of the metals. Ongoing computer modeling that considers only the former process will be reported soon. At that point, we should be able to better assess the importance of including cation exchange when predicting the migration of metals in acidic uranium mill tailings solutions percolating through sediments.

Unfortunately, data for radium were not reliable and do not allow us to speculate on its fate. In future laboratory studies, we will give special attention to determining the migration potential of radium.

A comparison of the data from the different column sizes (4.5 cm up to 65 cm) and different residence times per pore volume (40 days versus 0.75 days) shows favorable agreement. Therefore, we feel that the laboratory data and tools are applicable and useful for predicting the fate of contaminants at actual uranium mill tailings disposal sites.

In summary, the empirical trace contaminant data suggest that:

- arsenic, chromium, lead, silver, thorium, and vanadium are readily immobilized by the clay liner and not released after acid conditions prevail
- cadmium, iron, manganese, nickel, zinc, similar trace metals, and uranium are immobilized by the clay liner as long as the solution pH is not driven below pH 4.5 by continual acid solution attack
- portions of selenium and molybdenum are very mobile, and travel with the same velocity as the water
- laboratory results can be extrapolated to actual field disposal conditions.

We have identified other probable controlling mechanisms such as precipitation of hydrous oxides, formation of element-specific low-solubility compounds, anion exchange on protonated hydrous oxide surfaces, and cation exchange on the clay liner surfaces. Table 3 is a summary of the migration potential of selected macro anions, trace metals, and radionuclides based on the various types of empirical data presented in the text.

In addition, eight permeability columns containing coarse materials were set up and connected to syringe pumps. The syringe pumps insure that constant flow through the columns is maintained. We packed two of the columns with shaft-wall material from the UNC Churchrock mine in New Mexico. We also prepared duplicate columns with Lucky Mc Pathfinder overburden, overburden from the Exxon Highland Mill mine, and coarse shaft-wall material from the Morton Ranch mine. The columns were leached with ground water to establish a chloride-background baseline. Once the chloride baseline had been established,

TABLE 3. Summary of Migration Potential

Element or Constituent	Immobilized at all pH Values (3.5 - 8.5)	Immobilized at Near- Neutral pH Values (4.5 - 8.5) but Mobile at Low pH Values (pH <4.5)	Mobile	Insufficient Data
NO ₃			X	
Al		X		
SO ₄ ²⁻			X	
V	X			
Cr	X			
Mn		X		
Fe		X		
Co		X		
Ni		X		
Cu		X		
Zn		X		
As	X			
Se			X	
Mo			X	
Ag	X			
Cd		X		
Ba	X			
Pb	X			
Ra				X
Th	X			
U		X		

the columns were injected with chloride spikes, and effluent chloride concentrations were monitored to provide data to compute the dispersion coefficients for these coarse materials.

Task 3. Geochemical Modeling

An ion-speciation-solubility and mass transfer model (MINTEQ) developed at Pacific Northwest Laboratory (PNL) was recently made available for use in modeling the reactions in uranium mill tailings. Verification work was done in FY-82 to compare MINTEQ to other computer codes such as PHREEQE, WATEQ3, and EQUILIB. MINTEQ results have been consistent with the output of the other ion-speciation solubility and mass transfer codes that have been tested. The MINTEQ model has several user-oriented options. For example, the model will select which minerals or solids to dissolve or precipitate to achieve the most stable equilibrium state for the entire data base, or let the user restrict the number of phases from which the model will choose. It also allows the user to specify with which solids and minerals the solution should come to equilibrium. In addition, several adsorption options enhance MINTEQ's versatility when applied to uranium recovery operations. One can use everything from an empirical, K_d-type approach up to a more theoretically sophisticated, triple-layer, site-binding approach, depending on the amount of input data available.

The chemical reactions for Ra, Se, Mo, and Th along with their associated log K's have been compiled so that they may be added to MINTEQ. The reactions for these four elements have been coded into the model. We are currently evaluating the literature on Cr. Subsequent to this evaluation, Cr will be added to the data base. By increasing the data base, we improve MINTEQ's capabilities to model the chemistry of uranium mill tailings solutions. Internal consistency was maintained by ensuring that $H_{f,298}^0$, $G_{f,298}^0$, and S_{298}^0 satisfy the following expression

$$WG_{f,298}^0 = \Delta H_{f,298}^0 - T\Delta S_{f,298}^0.$$

We are actively using laboratory data from permeability columns 1 and 5 in geochemical modeling efforts. The columns contained the Morton Ranch clay liner leached with the synthetic tailings solution and the Highland Mill tailings solution. A total of 14 effluent solutions from these columns were modeled. These columns had been leached with their respective solutions for periods extending up to three years. Batch studies were also modeled where original Highland Mill tailings solutions, synthetic tailings solutions, and sulfuric acid were each separately reacted with: 1) a clay liner from Morton Ranch, Wyoming; 2) a sodium-saturated bentonite; and 3) a polymer treated bentonite. The tailings, synthetic tailings, and H_2SO_4 solutions were reacted for 16 months, 12 months, and 1 week, respectively. Modeling results were obtained for a total of 17 solution analyses.

Our conclusions from the calculated results of the ion speciation-solubility portions of the MINTEQ geochemical model are listed below.

1. The geologic materials with which the solutions were reacted have the capacity to mitigate the concentrations of possible contaminants in the solution via precipitation reactions.
2. Solid phases, such as gypsum and $Fe(OH)_3(A)$, may be precipitating in the reacted liners and result in decreases in the porosity and permeability.
3. Precipitation is observed to be a function of the liner/solution ratio with higher ratios having a tendency to precipitate more solids.
4. Higher liner/solution ratios have higher pH values which explains the greater precipitation noted in conclusions 3.
5. Amorphic metastable solid phases may control the solubilities of their dissolved constituents [e.g., $Fe(OH)_3(A)$ and $SiO_2(A)$].
6. Jarosites were computed to be supersaturated in the solutions resulting from the batch interaction of H. M. tailings solution with all geologic and liner materials and in all the column effluents where adequate solution analyses were available, and were identified by X-ray diffraction in both the column and batch experiments.

7. Solid phases identified as exercising possible controls on solution chemistry are $\text{Fe}(\text{OH})_3(\text{A})$, AlOHSO_4 , alunite, kaolinite, celestite, anglesite, gypsum, MnHPO_4 , and $\text{SiO}_2(\text{A})$. Solubilities of many elements and compounds were pH dependent (e.g., Fe, Al, Si and SO_4^{2-}).
8. For certain elements, such as Si and Al, different solubility controls may be operating at different pH values.

Certain solids identified as being in equilibrium with the solution chemistry were either poorly crystalline or present in such trace quantities that it would be impossible to identify their presence by X-ray diffraction.

It should be remembered that these results and conclusions represent an exploratory effort to geochemically model uranium mill tailings solutions, with their associated high sulfate concentrations, low pH values, and high total dissolved solids. The results and conclusions are based on data from a limited set of experiments. The solution analyses from these experiments were modeled as reported, without judgement on the reported concentrations, even though the cation/anion imbalance exceeded 20 percent in several modeled solutions.

These modeling results illustrate the paramount importance of obtaining accurate and adequate solution analyses. They also demonstrate the importance of close collaboration of modeler and experimentalist so that analyses crucial to the modeling effort are performed.

This exploratory use of geochemical modeling demonstrates the ability of MINTEQ to test solubility hypotheses for compositions of aqueous solutions that have high sulfate, low pH values, and high total dissolved solids, such as acidic uranium mill tailings solutions. Modeling can also guide the selection of important constituents and parameters that should be analyzed in these solutions. It may also point to thermodynamic data that may be in need of re-evaluation. Geochemical modeling can be used, in parallel with characterization techniques, to substantiate the presence or absence of solid phases in laboratory solubility studies. As used here, geochemical modeling is an effective tool to delineate the chemical causes for changes in permeability of clay liner materials.

From our studies, we concluded that the laboratory experiments and geochemical modeling predictions simulated field observations. The same mineralogical changes and contaminant reductions observed in the laboratory studies were found at a drained evaporation pond (Lucky Mc Pathfinder in Wyoming) with a 10-year history of acid attack. Geochemical (thermodynamic) modeling also predicted that the same minerals observed at the pond would form upon neutralization of the acidic tailings solutions by the contacted sediments. Thus, the laboratory studies, field observations, and theoretical model predictions complement each other.

We also began geochemical modeling studies using the mass transfer capabilities of the MINTEQ code. The code was used in a predictive mode to simulate the pH-front breakthrough and effluent compositions in both permeability and vacuum-extractor columns of uranium mill tailings percolating through the

Morton Ranch clay liner. Through this work, we attempted to validate the chemical model postulated for a system involving interaction of geologic sediments with uranium mill tailings.

Predictions were good for concentrations of several macro constituents (e.g., Fe, Al, Mn, and SO_4) of the tailings solutions, but were poorer where the trace metals were concerned. It appears that for several of the elements studied, other mechanisms (e.g., adsorption) become important factors in addition to the precipitation/dissolution and speciation reactions considered.

PROJECTED WORK FOR NEXT YEAR

Efforts in FY-84 will concentrate on field site surveys to collect necessary and sufficient geochemical and hydrologic data on ground-water, tailings, tailings solutions, liners, and overburden materials to verify past laboratory and geochemical modeling predictions. Our field-site surveys will involve sampling soil pore waters at different depths beneath the pond to determine the concentrations of various contaminants with regard to distance away from the mill pond. Again, we will use the data to verify geochemical modeling predictions and laboratory experiments. We will also perform any ongoing or additional laboratory work that is needed to facilitate the comparison of modeling and field data. All planned laboratory work will be completed in FY-84.

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

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OBJECTIVE

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

ACCOMPLISHMENTS DURING REPORTING PERIOD

Task 1. Project Management

During the past year, background information on aquifer restoration applicable to in-situ uranium mining was reviewed and compiled in a project document (NUREG/CR-3104). We designed and carried out experiments on natural restoration processes using sediment and lixiviant samples that were collected at a mine site in south Texas. The results of these experiments were published in a technical progress report (NUREG/CR-3136). We presented two speeches on the results of our field and laboratory work at meetings of professional societies and the texts of those presentations will be published in the proceedings of the symposia. The initiation of laboratory experiments on induced restoration techniques was planned and screening tests have begun. Details on our technical accomplishments may be found in the appropriate task descriptions. The reports and published articles are listed in a separate section. Additional details on project management activities follow.

The original scope of this project included a study of excursion monitoring methods and an assessment of the long-term impact of lixiviant excursions on ground-water quality. The task that included these aspects of in-situ uranium mining was eliminated from the project near the end of FY-82 because of reduced funding. However, interest in excursion identification and control has grown in the last year, and we are developing a new test plan to study these factors. A research proposal to perform the excursion studies is being prepared.

As a result of interest that was shown at the February 1983 project review, we made an additional change to the scope of the current in-situ

project to include sampling of a Wyoming in situ mine site. In order to show similarities and differences that might occur during aquifer restoration in Texas and Wyoming, funds have been allotted to sample a Wyoming site and perform a geochemical characterization study of the ground water and sediments. The results of this characterization will be compared to similar work done on the Texas samples.

Because of their experience with in-situ mining techniques and their knowledge of the uranium mining industry, Daryl Tweeton and his coworkers at the Twin Cities Research Center of the Bureau of Mines were hired as subcontractors to this project. During the past year, they provided input to the document on aquifer restoration regulations and techniques (NUREG/CR-3104), and compiled baseline geologic and geochemical data for in-situ mine sites in the state of Wyoming. Currently, they are conducting laboratory experiments on restoration to complement those being done at PNL.

Task 2. Site Characterization

The ground water, leaching solution (lixiviant), and sediments collected during March 1982 at the Benavides (Texas) mine site of Uranium Resources, Inc. have been analyzed and the data have been interpreted as they apply to aquifer restoration and excursion detection. The Bureau of Mines has compiled data for us on several mine sites in Wyoming to compare to the Texas operations. The details on this work follow.

The lixiviant sampled at the Benavides site contains high total dissolved solids (TDS >4,000 ppm) compared to the ground water (average TDS <1,500 ppm). The concentration of each major cation and anion is greater in the lixiviant compared to the ground water; however, the ratio of concentrations varies significantly by element. We computed the ratio of major ion concentrations in the lixiviant and ground water by using average concentration values for the two production wells sampled and representing the ground-water composition of the ore-zone aquifer as the average of ten wells. Table 4 lists these average concentrations and the computed ratios. It shows that the concentrations of Ca, Mg, and SO_4 in the lixiviant are highly elevated compared to those in the ground water. The lixiviant used at this leach facility is ground water fortified with oxygen and carbon dioxide gas, neither of which would directly add Ca, Mg, or SO_4 to the solution. The large rise in concentration of those components results from interactions between the lixiviant and sediment that include oxidation-reduction and dissolution of the aquifer minerals.

The trace metal content of the lixiviant and of the ground waters sampled from the Benavides site is generally very low, if not below detection limits. The concentrations of As, Ba, Cd, Cr, Pb, Ag, Cu, Fe, and Zn do not exceed EPA primary or secondary drinking-water standards. Selenium is present in the two lixiviants that we sampled at 0.018 and 0.028 ppm. These levels exceed its allowable maximum concentration level (MCL) of 0.01. However, selenium is also present in some of the ground water (Well 831) at a concentration of 0.48 ppm; therefore, mining has probably not generated the hazard.

The bulk chemistry and mineralogy of the reduced zone sediments at the Benavides in-situ leach facility have been determined. The site was characterized to establish the baseline condition, and thereby provide a reference

TABLE 4. Mean Concentration and Standard Deviation of Selected Constituents in the Ground Water and Lixiviant

	Ground Water		Lixiviant		Concentration Ratio Lixiviant/Ground Water
	x, ppm	σ	x, ppm	σ	
Ca	25.9	14	273.0	61	10.5
Mg	9.6	5.8	82.0	10	8.5
Na	486	90	1007	89	2.1
K	15.2	2.3	26.5	11	1.7
Cl	677	196	1009	33	1.5
SO ₄	71.1	15	1181	131	16.6
HCO ₃	173	36	579	79	3.3
pH	8.53	0.24	6.71	0.02	1.3 (log units)
U	0.17	0.21	28.6	36.4	168

that we can compare the laboratory restoration experiments against. Routine optical microscopy results indicate that feldspar and quartz dominate the sand comprising the ore-zone aquifer with less than 5 percent opaque minerals. Because sulfide minerals have an important role in both ore genesis and restoration processes, a heavy mineral separation was made. X-ray diffraction patterns of both the light and heavy fractions indicate that plagioclase feldspar and quartz dominate the light fraction, while the heavy fraction is principally pyrite with minor marcasite and ilmenite.

We prepared a clay separation from the bulk sample to identify the type of clay minerals present. Oriented samples of the clay fraction (estimated at 5 percent of the bulk sediment) were prepared and treated by standard clay mineralogy procedures. Diffraction patterns of the treated clay indicate a montmorillonite with possibly a trace of illite. X-ray photoelectron spectroscopic analysis of the clay fraction shows the montmorillonite to be a sodium-rich variety.

The mineralogy of the major constituents of the South Texas Benavides ore-zone aquifer is similar to that found at several Wyoming in-situ sites. The data for the Wyoming sites were compiled by the Bureau of Mines as part of their work for this project. The in-situ mining operations that were used for the compilation are the Bison Basin Project (Ogle Petroleum, Inc.), Collins Draw Project (Cleveland Cliffs Iron Company), Highland Project (Exxon), Irigaray Project (Wyoming Mineral Corporation), and Nine Mile Lake Project (Rocky Mountain Energy Company). In most cases, the host medium for the ore

is an arkosic sandstone composed predominantly of quartz and feldspar with minor clay, mafic minerals (hornblende, pyroxenes, and biotite), and organic matter. Calcite often occurs as a cementing agent for the sands. Uraninite is the typical ore mineral, however coffinite is sometimes reported associated with the uraninite in the mineralized zone. Selenium and arsenic-containing minerals were reported at two of the Wyoming sites, and their occurrence could cause restoration complications. Selenium was reported to be present as native selenium and in the mineral ferroselite at the Highland Mine site, and arsenic was reported as a component in barite found at the Irigarary site. The solubility of arsenic and selenium minerals is redox-sensitive, consequently, leach mining could mobilize these elements.

Uranium remaining in the sediments following leaching may also be dissolved by ground water or residual lixiviant. To ascertain whether this poses a significant problem, we determined the uranium content of leached ore from one of the core samples from the Benavides mine. The section of the ore zone from which the core was taken had been actively leached for approximately two years. Company records show that at the commencement of mining the average uranium content of the nine-foot thick ore zone was 0.16 weight percent (as U_3O_8). The uranium content of the eight leached samples that we analyzed ranged from 0.003 to 0.014 weight percent. Although an appreciable quantity of uranium had been removed from the zone, a significant amount (up to 140 ppm) remains as part of the sediment. Because of decreasing recovery, this leach area will be closed soon. Restoration plans for this leach field should take into account the possibility of additional oxidation and mobilization of uranium left in the sediment.

Task 3. Natural Restoration

During the past year, we performed two sets of batch and flow-through column experiments to evaluate the effectiveness of natural processes in restoring aquifer chemistry to pre-mining conditions. We used samples of ground water, lixiviant, and aquifer sediment from the Benavides mine in these experiments. The first set of experiments using both batch and column methods was conducted with the equipment exposed to the atmosphere. We noted very little long-term effect of solution/sediment interactions on the redox potential or composition of the solution. This suggested that oxygen from the atmosphere may be affecting the restoration capacity of the sediment. The second set of batch and column experiments were performed in a controlled atmosphere (argon) chamber in an attempt to decrease the chance of oxygen contamination, and thereby more closely simulate the deep aquifer reducing environment that is associated with the uranium ore. The experiments are described in detail in NUREG/CR-3136 and the results of the second set of experiments are summarized below.

In the controlled atmosphere chamber, we were able to keep the oxygen level in the atmosphere at approximately 110 ppm, which is well below the 200,000 ppm level of the open atmosphere. During the batch experiments, we monitored the pH, Eh, and dissolved oxygen content of the solutions that were obtained by mixing lixiviant and aquifer sediments. In all cases, the dissolved oxygen level decreased from 8 ppm (equilibrium with the open atmosphere) to 0.1 ppm (less than or equal to the detection limit of the oxygen

meter). The pH varied slightly in response to purging the solution of CO₂ gas and equilibration of the solution with calcite in the sediment. The redox potential, Eh, of the lixiviant responded to gas purging of the solution by dropping from +300 mv to +200 mv. When sediment was added to the solution, Eh dropped an additional 100 mv soon after addition of the solid. However, monitoring the Eh of the lixiviant/ sediment mixture over the period of a few hours showed that the Eh slowly rose back to +200 mv. The experiments were carried out in beakers open to the atmosphere of the agron chamber, and even though the oxygen content of the chamber is low, it apparently is enough to influence the Eh of the solutions.

The composition of the solutions obtained from the batch experiments suggest some of the geochemical interactions that occur between the lixiviant and the aquifer sediments. In most tests calcium concentration increased over the first few hours and then either remained stable or decreased to an intermediate value. Carbonate concentration, on the other hand, decreased by 50 percent throughout the lixiviant/sediment experiments. The increase in Ca concentration may result from desorption of Ca from surfaces of the sediment minerals in response to the introduction of the Na-rich lixiviant. Sodium in the ground water from the ore zone has a mean concentration of 484 ppm whereas the lixiviant has a concentration of approximately 1,000 ppm. The mixing of lixiviant with sediment would result in ion exchange of Na for Ca on the mineral surfaces, an increase in Ca concentration in the solution, precipitation of calcite, and a resulting decrease in carbonate concentration.

In summary, the one and two-day batch experiments showed that the sediments can affect the redox potential, dissolved oxygen content, and composition of the solutions they contact. However, if the solutions are in contact with an atmosphere that contains as little as 100 ppm oxygen, the effect of the sediment on the Eh of the solution will be modified. This may also affect the composition of the solution. The lessons learned from the batch tests were used in designing the column experiments which are described next.

In the column experiments, lixiviant was pumped through acrylic plastic tubes filled with aquifer sediment. We monitored the pH and Eh of the effluents by means of an in-line flow cell, and collected the solutions for chemical analysis. The entire apparatus was kept inside a controlled atmosphere chamber during the experiment. Our results show that the redox potential (Eh) and the concentrations of uranium and carbonate in the lixiviant are greatly affected by contact with a small amount of sediment. The Eh of the lixiviant changed from +300 mv to -300 mv; the uranium concentration dropped from 52 ppm to less than 5 ppm; and carbonate concentration decreased by half. The concentration of most of the major cations (Na, Ca, Mg, and K) and anions (Cl and SO₄) in the column effluent was equal or close to that of the influent lixiviant. This suggests that the major constituents of the lixiviant are not significantly retarded by chemical reactions as they pass through the column.

Our experiments showed that the dissolved oxygen level of the solution is effectively lowered by water/sediment interactions. Uranium will precipitate in the columns because the redox condition changes; we expect a similar response from the redox-sensitive trace metals (e.g., As, Se, and Mo). The carbonate

concentration changes in response to the precipitation of calcite. Carbonate minerals may scavenge trace metals from solution as they precipitate; this process would also lower the solution concentration of these metals which are potential byproduct contaminants of uranium leaching. Because some of the dissolved constituents (e.g., Cl and SO₄) of the lixiviant are not affected by flow through the columns, we believe that these constituents, as well as the total dissolved solids level in the water, would be good excursion indicators for most mine sites.

Task 4. Restoration Techniques

We are using the results of the experiments on natural restoration and our practical experience in working with lixiviant and ore material to design experimental methods for investigating induced restoration techniques. The applied techniques that have been used in the past or that have good potential as a restoration method include water recirculation with chemical additives and recirculation with surface treatment (e.g., reverse osmosis and electro-dialysis). During FY-83, we have begun studying the applicability and effectiveness of chemical addition techniques to aquifer restoration, and in FY-84 we will evaluate surface treatment methods.

We have established a tentative schedule of experiments designed to evaluate the potential for chemical injection and recirculation techniques to enhance aquifer restoration. Currently, we are analyzing thermodynamic and kinetic data to choose reductants for laboratory testing. Batch tests with selected reductants and ground water and lixiviant will be carried out to determine the compatibility of the reductants with the solutions. This will be followed by a series of batch tests with solutions in contact with leached ore and reduced sediment. We will complement the batch tests with flow-through column experiments where lixiviant with the reductant added, will flow through sediment.

Task 5. Field Test Assessment

A small-scale field demonstration of the restoration techniques developed in the project may be necessary to test the techniques and show commercial applicability. According to the latest Statement of Work, the plan for the field test will be written after obtaining approval from the NRC project manager. We expect that a decision on the necessity for a field test can be made early in FY-84.

PROJECTED WORK FOR NEXT YEAR

During the coming year, an in-situ mine site will be sampled in Wyoming, and the characteristics of that site will be compared to the Benavides site in Texas to determine commonalities and differences between the sites that might affect restoration. To the extent that funding is available, we will perform laboratory experiments on the sediment and water samples to study natural and induced restoration.

The natural restoration experiments begun previously on the Texas samples will be extended to evaluate the potential for natural processes to reduce

trace metal contaminants (e.g., As, Se, and Mo) from the ground water. We will conduct a series of batch and column experiments to test the effectiveness of the selected chemical additives to restore the ground water. We will include the results of these initial tests on induced restoration in a letter report to be submitted by November 1983. The results of widespread industry efforts to restore ground water by surface treatment of the residual solution in the leach field will be compiled and evaluated. In addition, other promising techniques for restoration will be evaluated and described in the final technical report for this project.

If the NRC decides that a field test of restoration techniques is warranted, a test plan will be written. The test plan will include scope, technical approach, and specific task descriptions focused on the field site.

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

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RELATED RESEARCH ON URANIUM RECOVERY

Related research programs in Uranium Recovery (UR) include the following U.S. and International Activities:

RELATED RESEARCH IN THE USA

The other major research effort in the USA has been by the U.S. Department of Energy (DOE) Uranium Mill Tailings Remedial Action Program (UMTRAP). The UMTRA Project Office of the U.S. Department of Energy (DOE) has supported a multi-faceted technology development program to assist in the performance of remedial action at 24 inactive uranium mill tailings piles. The work under the UMTRA program was initiated in FY-80 and the majority of the work is scheduled for completion in September of this year. The major areas of research and development in this program are outlined below:

Cover Technology

- Asphalt Emulsion Admix
- Earthen Covers (Single and Composite)
- Revegetation/Riprap
- Biobarriers

Liner Technology

- Liner System Studies
Asphalt and Clay Liners
- Grouting Technology

Measurements/Monitoring

- Radiological Data
- Instrumentation
- Technical Measurement Center at Grand Junction, Colorado

Tailings Conditioning/Mineral Recovery

- Assay/Reprocessing
- Reconditioning

Hydrogeochemical and Geotechnical Studies

- Hydrology
- Geochemical Studies
- Geotechnical Studies

A summary of research highlights is included in the following section.

Cover Technology

The cover system is designed to limit radon emanation rates and to prevent plant or animal intrusion or erosion. Several designs have been tested in this program.

Asphalt Emulsion Admix

An asphalt emulsion sealant of approximately 7 cm thickness is being studied by Battelle Pacific Northwest Laboratory (PNL) as a radon barrier. After the asphalt emulsion admix is applied and compacted on top of the tailings, a 0.6 - 1.0 meter layer of overburden is applied to protect the seal from ultraviolet degradation. Soil is then placed on top of the overburden. We use an admix seal of cationic asphalt emulsion and a fine aggregate such as concrete sand to establish an electrostatic bond, which forms a continuous membrane. The major concern regarding the use of an asphalt emulsion is its long-term stability in the light of possible subsidence, oxidation, microbial attack, and freeze-thaw cycles. Four years of laboratory and field tests have indicated that the asphalt emulsion can effectively prevent radon diffusion. An extensive and large-scale field experiment was initiated in 1981 at the Grand Junction tailings pile in Colorado to provide better answers to the questions regarding the feasibility of an asphalt-emulsion cover. Different application techniques have also been tested, and the cold mix paver sealant seems to result in the best seal. This seal meets the new EPA standard of $20 \text{ pCi m}^{-2} \text{ s}^{-1}$, and also appears to be an effective biobarrier and long-term cover.

Earthen Covers

Both single and composite earthen cover systems have been tested. The single-layer system consists of a "dump and spread" soil cover of sufficient thickness to reduce the radon flux to an acceptable level. A three-meter thick, single-cover system is being tested at Grand Junction by PNL. Additional tests on earthen cover are also being made at Grand Junction that will reveal the effects of compaction, thickness and moisture content on radon exhalation. Data from this test effort will be used designing of radon-control cover systems.

A multilayer cover system that could be used in place of thicker one-layer systems is also being investigated by PNL and Rogers & Associates. The layering sequence for this system consists of a wet clay layer placed on top of the tailings, a coarse layer applied on top of the clay, and fill and topsoil placed on the top coarse layer. The clay layer remains saturated due to the hydraulic barrier that is established, and the top coarse layer acts as the capillary barrier. This barrier is a layer of clean, washed rock that isolates the wet clay from the topsoil. In addition to keeping the clay layer wet, the barrier limits the upward migration of contaminants from the tailings. The moist clay acts as a radon seal and less cover thickness is required to meet the EPA radon exhalation standards. The total thickness of the cover system is approximately 1 1/2 meters as compared to the referenced three meters for a single-layer earthen cover. The large-scale field test at the Grand Junction tailings pile includes a multilayer cover system with wet clay mixtures. The large-scale experiment provides excellent side-by-side comparison of the performance of the asphalt seal, the multilayer and earthen covers.

Revegetation/Riprap

The studies in this task are concerned with schemes to revegetate, and thus stabilize the topsoil covering the tailings impoundment system. The types of species, nutrient requirements, how and when to plant, the root depth, erosion potential, and long-term stability of the plants are being investigated. The riprap task examined different possible rock covers and the interaction between plants and rocks. A modeling and optimization effort is also being pursued in this area. The methodology being developed will determine whether riprap or vegetation, or a combination of both, is preferred for a specific disposal site. The cost for vegetation and rock cover systems will also be determined. In addition, the effect of surface stabilization treatments on moisture retention in the covered piles is being examined. While vegetation can result in a stable moisture content, the evapotranspiration from rock cover systems can cause drainage due to an evaporation reduction and an increase in moisture content. This can, in turn, result in a ground water pollution problem.

Biobarriers

The use of physical barriers to control animal intrusion and prevent root growth into the impoundments We studied a coarse rock layer was found useful as an animal and possibly a plant root barrier, and a chemical herbicide in the form of pellets was shown to prevent root intrusion into the seal. While a pellet system lasting 100 years was attainable, a 1,000 year pellet system is beyond our reach at this time. Thus, gravel or asphalt barriers were recommended. For gravel barriers, there appears to be a direct relationship between the rock size in the layers of loose rocks that are placed below the soil surface, and the size of the animals which are deterred from penetrating the radon seals.

Liner Technology

The investigation of different liner materials by PNL and Oak Ridge National Laboratory consists of evaluating the effectiveness of liners and evaluating grouting techniques. Accelerated aging tests of different materials have been performed, and field tests of suitable liners are underway at the Grand Junction pile. The most suitable liner materials have been determined to be asphalts, natural soil and clays. Of the eight liner materials that have been tested, catalytic airblown and asphalt and soil amended with 10% sodium bentonite were revealed to have superior liner characteristics. Criteria are being developed that will be used to determine whether a liner is necessary on a site-by-site basis. The design parameters for liners will also be defined. Assessment of grouting techniques by Oak Ridge indicates that for sites that may require moving, grouting may be a feasible and economically sound alternative.

Measurements/Monitoring

This work has included obtaining and evaluating data describing radiation and radioactivity levels at the sites, implementing radiological surveillance

programs to provide the required data before and after remedial action at all the sites, and developing models that predict radiological impacts from the storage and transportation of uranium mill tailings. Also, an effort to coordinate radon and radon-daughter instrumentation and measuring techniques was begun in 1982. While Argonne National Laboratory has been extensively involved in this program, Bendix has been designated as the measurements center and will coordinate this particular effort. Additional discussion of this effort is given below.

Technical Measurement Center

The Technical Measurement Center (TMC) was established by the DOE Division of Remedial Action Projects in support of the UMTRA Program, the Grand Junction Remedial Action Program (specific for radiological cleanup of offsite dwellings and buildings in and around Grand Junction, Colorado), the Formerly Utilized Sites Remedial Action (FUSRAP) and the Surplus Facilities Management Program. Bendix (Field Engineering Corporation) was selected as the contractor for the Technical Measurements Center (TMC) and is located in Grand Junction, Colorado. Work assigned to the TMC include the following environmental monitoring efforts and protocol development tasks:

- The development of protocol for gamma-ray measurements for radium and for whole body exposure rates. (A draft protocol has been developed in FY1983.)
- A report on alternatives to an annual average working level measurement.
- Establishing a working level (WL) calibration facility. This effort is underway; expected to be complete in September 1983.
- Intercomparison and calibration of radon measurement instrumentation. Instrument testing is underway.
- Developing a protocol for time-integrated measurement of radon concentrations.
- Evaluation of instrumentation for measuring radon-daughter.
- Developing a protocol for collecting and preserving field samples (soil and water sampling).
- Developing a protocol for analytical field methods.
- Development and testing of appropriate calibration test facilities.
- Procurement of a calibrated Ra-226 source for instrument calibration.

Reprocessing/Reconditioning/Mineral Recovery

Reprocessing

Through fiscal year 1981, ORNL looked at actinide (Ra, Th) separation of the tailings by nitric acid leaching. This effort was discontinued due to several problems regarding this process; it is not a heap leach process because it must be performed at high temperatures, special equipment is required, and

nitrate which can cause water contamination are produced. Los Alamos National Laboratory (LANL) has evaluated reprocessing using concentrated sulfuric-acid leaching, which can remove the radionuclides and residual minerals. Substantial recovery of minerals requires strong acids, and concentrated sulfuric acid can extract significant percentages of radium-226, barium, thorium-230, uranium, molybdenum, cobalt and vanadium.

Reconditioning

Los Alamos National Laboratory (LANL) investigated the immobilization or removal of the contaminants through two main activities. The main focus of the studies was thermal stabilization, whereby the tailings were heated to 1200 C to radically alter the tailings structure. The sintering process forms a slag, thereby reducing the radon emanation. Mineral transformations were shown to take place, and up to 99% of the radon was reduced. The slimes were turned into glass and the sands turned into aggregate crystals. A rotary cement kiln was used to sinter the tailings. An economic analysis of this concept has indicated that it can be competitive at certain tailings sites where the other remedial action option is transporting the tailings to a new disposal site.

Assay/Reprocessing

The feasibility of reprocessing the tailings at 13 of the 24 sites is being investigated by means of assays, tests and economic evaluations. EPA listed elements are being identified in the tailings, and the radium concentrations beneath the piles and in the storage pond areas are being determined. Furthermore, as an adjunct of the assay test program, additional data is being gathered to support sections of the technology development program. For example, the engineering properties are being determined and piezometers are being emplaced to evaluate the release mechanisms and transport phenomena of the contaminants. Sandia Laboratories, Mountain States Mineral Enterprises, Sergeant Hauskins and Beckwith, and Colorado State University (CSU) are involved in these activities.

Geohydrochemical and Geotechnical Studies

Hydrogeochemistry

A hydrology effort has been initiated by Lawrence Berkeley Laboratory (LBL), with assistance from CSU, to look at radionuclide release mechanisms and water table contamination. Two sites (Riverton and Maybell) are being looked at in detail, although some data has been gathered at all of the 13 sites being studied in the assay program. A hydrology model which addresses contaminant-mobilization mechanisms and transport rates is being tested by LBL.

Geochemistry

A geochemistry program consisting mainly of detailed chemical analysis at key UMRAP sites is being pursued at Geochemistry and Environmental Chemistry Research, Inc. (GECR). This program has developed site-specific characterizations of the geochemical nature of the tailings and the extent of the contamination at each site. The program has assessed the magnitude and extent of contamination

at the sites, examined the mobility of contaminants, and assessed the significance of chemical conditions and reactions which may affect the stabilization systems. The initial conclusions by GEGR are that contamination is mainly upward and toward the surface for tailings piles in dry environments. Thus, according to GEGR, there is little need for liners at most sites, and chemical processes must be examined in order to locate suitable disposal sites.

Geotechnical

A tailings stability program is underway at Colorado State University (CSU). CSU will issue site reports to describe the engineering properties, such as moisture, density, grain size distribution, compaction, and permeability of the tailings at all sites.

A very small carry-over budget is anticipated for the technology development program for FY-84, and no further research effort is planned by the UMTRA Program after FY-84. The status of the TMC past FY-84 is not known. A list of the remedial action sites is included for reference and indicates that action has been taken to start remedial programs in early FY-83 at Canonsburg and Salt Lake City.

CANADIAN RESEARCH

Canada, with 130M tons of uranium tailings covering more than 10 square kilometers, is seriously concerned about uranium mill tailings. The Canadian government has recently established a multimillion dollar research program on uranium tailings. The program is directed by the Canadian Department of Energy, Mines and Resources (CANMET). In Canada site conditions are primarily wet, so water borne effluents (radium and toxic metals) are the source of environmental contamination from the existing major tailings piles. To date, little effort has been expended on studies of radon gas migration.

Research efforts include the study of heavy metal and radium migration from oxidized, pyritic tailings. Also under study is deep lake disposal of tailings as well as physical/chemical stabilization of tailings piles using commercial stabilizing agents.

The major objectives of the Canadian research program are similar to those in the U.S., and address the long-term management of tailings, and optimal disposal methods to ensure minimal migration contaminants. Several contacts have been made with Canadian researchers regarding specific research activities including the new uranium mill tailing (UMT) program directed by CANMET. Mr. Vic Haw, director of the Canadian UMT program is planning a visit to PNL this summer and is prepared to assist in information exchanges between Canada and the U.S.

AUSTRALIAN RESEARCH

Australia, having produced over 135M metric tons of uranium tailings, also has a significant interest in research related to uranium tailings management. Current uranium-mining activity in Australia is mainly centered in the Alligators River Region in the Northern Territory. Research efforts are concentrated on disposal of tailings in this region. The Australian government has imposed the concept of "Best Practicable Technology" on the management of uranium tailings. This concept requires that technology be used in mill tailings management that produces the minimum amount of environmental pollution and degradation that can "reasonably be achieved." Specific regulations are still being developed and the Australian government is funding a modest program in uranium mill tailings research through the Environmental Chemistry Section of the Australian Atomic Energy Commission. Studies of process mechanisms for geochemical transport, radium chemistry and field experiments on radium uptake by plants are currently part of the Australian research program.

INTERNATIONAL ATOMIC ENERGY AGENCY (IAEA) COORDINATED RESEARCH PROGRAM ON RADIUM MIGRATION

The IAEA is presently sponsoring a coordinated research program (CRP) on "Environmental Migration of Radium and Other Contaminants Present in Liquid and Solid Wastes from the Mining and Milling of Uranium." This research program is focused on radium migration from uranium mill tailings and includes participation from Argentina, Australia, Brazil, Canada, Czechoslovakia, India, Japan and the United States. This program is an extension of a previous program on radium migration and was initiated for a 3 year period in May 1982. Exchange of research

information on radium migration, particularly between research groups in Australia, Canada and the U.S. is expected to be beneficial to the NRC (and PNL) research efforts. A meeting of the CRP in Brazil has been tentatively scheduled for early 1984. The PNL research program, "Leachate Assessment" (B2292) has been designated as the official research program and the reports from this and other related programs at PNL will be exchanged with the members of the CRP.

ORGANIZATION FOR ECONOMIC COOPERATION AND DEVELOPMENT, NUCLEAR ENERGY AGENCY
(OCED NEA)

The NEA has recently completed a two volume report entitled, "NEA Study on the Long-Term Aspects of the Management of Wastes from Uranium Mining and Milling; Vol. 1 - "Radiological Aspects" and Vol. 2 - "Technical Aspects." This NEA report was prepared by the NEA Coordinating Group on Long Term Management of Uranium Mining and Milling Wastes. The U.S. Dept. of Energy (DOE) has been represented on NEA Working Groups by PNL personnel involved in the UMTRAP program and they and other UMTRAP participants reviewed this document in early February of this year. The report documents radiological effects from three hypothetical mill sites located in the U.S.A., Canada and Australia relative to ICRP (International) criteria and provides a generalized overview of the necessary technology for proper waste management.

In addition to the above report, the NEA, under the direction of Dr. A. Muller, is planning to produce several monographs specific to Uranium Mill Tailings. These monographs will provide detailed technical discussions of issues related to mill tailings stabilization. PNL has been contacted and asked to participate in contributing to a monograph on "Current Understanding of Geochemical Processes related to Uranium Mill Tailings Management." Discussions on seepage chemistry, tailings-sediment interactions, and transport model developments are planned. Research findings from the NRC program on Leachate Assessment (B2292) are planned to be included in this monograph.

PUBLICATIONS IN RELATED URANIUM RECOVERY RESEARCH

The following is a partial list of the published reports and papers available from the UMTRA project.

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