Uranium Mill Tailings Quarterly Report for the Period January-March 1982

**Compiled by
J. M. Latkovich**

May 1982

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Richland, Washington 99352

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JANUARY, FEBRUARY, MARCH 1982
QUARTERLY REPORT

RADON BARRIER SYSTEMS FOR URANIUM MILL TAILINGS

The overall objective of this project is to develop radon barrier systems that will meet EPA standards, minimize the use of remedial action, and remain stable for a very long period, including development of the supporting technology and engineering specifications.

SUMMARY

- Asphalt emulsion admix seal core samples taken from the 1981 field test site were characterized including radon diffusion coefficients, asphalt content and aggregate particle size distribution.
- Chemical stability studies continued including aqueous leaching and oxidation of asphalt seals and emulsion. After 1.5 yr of continuous exposure to brine solutions and deionized water no degradation has occurred.
- A range of soil types of 5 sites (Durango, Grand Junction, Riverton, Salt Lake City and Shiprock) have been characterized for properties that can be used to estimate radon diffusion through earthen cover systems. These properties include water retention characteristics, grain size analysis and bulk density. Field water content measurements with depth are useful in predicting expected long-term moisture contents of these soils.
- A pore size distribution model was used successfully to describe radon diffusion through soil cover systems over a wide range of water contents and material types.
- Both water drainage characteristics and sieve analysis were successfully used to predict pore size distribution and hence the radon diffusion characteristics of the soil materials tested.

- Three dimensional radon diffusion modeling was used to predict effects of cover defects on surface radon fluxes.
- A comparison between two dimensional numerical and analytical models of radon diffusion were found to be in agreement within one percent when run on a common test case.
- Cover placement on a drained tailings pile was modeled. A two dimensional radon profile was computed as a function of time before and after cover placement.
- Field data including moisture content, capillary pressure and temperature profiles from earthen covered plots at Grand Junction were analyzed. Frost penetration did not occur below a depth of 1 m. Moisture content and capillary pressure changes were small during the monitoring period (November through September 1981) but the data suggested that some drying of the wet clay occurred in the earthen cover column tests.
- Radon flux measurements were completed in February on the 1980 and 1981 field test plots including the asphalt emulsion radon barrier multilayer covers and earthen covers. Radon concentration profiles in the column were also determined.
- The 1981 Grand Junction field test costs were revised. Cost estimates for the earthen and asphalt radon barrier systems have been completed for Durango, Shiprock and Vitro sites. The cost of a multilayer cover will be calculated next quarter.
- Petroleum Sciences Inc., completed over 100 runs in developing the mix design test apparatus.

LABORATORY STUDIES

Asphalt Emulsion Barrier

Chemical Stability--D. A. Nelson

Laser-Raman spectroscopy was used to monitor the ratio of oxygen and nitrogen over asphalt samples sealed in glass ampoules. Two series of six

asphalt samples were examined; one through 60 days and another through 40 days. Reproducibility was achieved with one sample (asphalt emulsion seal applied 7/80 and collected 4/81). The remaining five samples agreed with the general trend of oxygen absorption for the respective material. Thus, older samples tend to absorb oxygen at a slow rate, while newer samples absorb oxygen at a more rapid rate. There is one definite anomaly to this presumed trend as noted in Table 1.

In both series of data, asphalt from the 7/80 seal-collected 4/81-absorbed oxygen at a rate much more rapidly than any seal samples from 7/81. This might indicate a larger amount of sulfur present in the 7/80 asphalt emulsion or some other unrecognized reactivity. We assume that sulfoxides are the first type of oxidation product formed. We will attempt to examine sulfoxide content in these asphalt samples. We also will reexamine the spectroscopic technique in order to achieve better reproducibility.

The asphalt samples were examined by differential infrared spectroscopy to determine the amount of carbon oxidation products. The results are present in Table 2. The ir procedure indicated that a definite buildup of oxidation products were observed with asphalt that was exposed to sun and air for 38 yr. However, no evidence, within detectability levels, was noted for oxidation in any of the newer asphalt samples.

TABLE 1. Laser-Raman Spectroscopy for Air Components Over Asphalt in Sealed Ampoules

#	Asphalt Type	O ₂ Absorption ^(a)
72A	7/81 seal (collected 2 days after laydown)	moderate
72B	7/80 seal (collected 4/81)	fast
72C	7/81 emulsion (from truck, not exposed to light)	moderate
72D	1946 asphalt (camp Hanford sidewalk)	slow
72E	7/81 seal (collected 10/81)	moderate
72F	same 7/81	moderate

(a) Fast indicates ~2 cm³O₂/g asphalt, moderate indicates 0.75-1.0 cm³O₂/g asphalt, and slow indicates ~0.5 cm³O₂/g asphalt. All values compared at 40 days exposure in sealed ampoules.

TABLE 2. Differential Infrared Spectroscopy of Asphalt(a)

#	Asphalt Type	Ketones	Oxidation Products (moles- ℓ^{-1})		
			Anhydrides	Free Acids	2-Quinolenes
72A	7/81 seal (collected 2 days after laydown)	0	<0.002	<0.02	0
72B	7/80 seal (collected 4/81)	0	<0.002	<0.02	0
72C	7/81 emulsion (from truck)	0	<0.002	<0.02	0
72D	1946 asphalt (Camp Hanford sidewalk)	1.2	0.12	0.11	0
72E	7/81 seal (collected 10/81)	0	<0.002	<0.02	0

(a) Procedure adopted from J. C. Petersen and H. Plancher, Anal. Chem., **53**, 786 (1981).

Thus, oxygen may be absorbed by these asphalt samples under ambient conditions (no intense ultraviolet light), but carbon oxidation has not been observed. We must assume that asphalt oxidation under these conditions is very slow.

Radon Diffusion Measurements--H. D. Freeman

Tests to determine the effective diffusion coefficient for radon through asphalt admix seals were conducted on cores from the 1981 Grand Junction asphalt seal; a foamed asphalt seal prepared by Ford, Bacon, and Davis, Utah (FBDU) and two pure asphalt cement specimens. The radon diffusion tests were run for 3 to 4 days for all test specimens. The effective diffusion coefficient was calculated from the collected radon that diffused through the seal and the measured below seal radon concentration by the transient diffusion model SEARCH. The results of the diffusion coefficient calculations are presented in Table 3.

After the cores were tested, each was extracted with Trichlorethylene to determine its asphalt content and the particle size distribution of the aggregate. The seven Grand Junction seals averaged $19.8 \pm 2.0\%$ on a dry basis (weight of asphalt/weight of aggregate). An attempt to correlate asphalt content to the effective diffusion coefficient resulted in Figure 1. Although

TABLE 3. Radon Diffusion Coefficients for Asphalt Emulsion Seal Core Samples and Other Asphalt Test Samples

Sample	Description	Diffusivity cm^2/s
1	1981 Field Test Asphalt Seal Core	4.93×10^{-6}
2	" " " " " "	3.19×10^{-6}
3	" " " " " "	4.40×10^{-6}
4	" " " " " "	2.83×10^{-6}
5	" " " " " "	1.05×10^{-5}
6	" " " " " "	2.22×10^{-6}
7	" " " " " "	5.02×10^{-6}
5B	Foamed Asphalt FBDU	1.05×10^{-5}
Cement 1	Pure Asphalt Cement Zero Pressure	4.36×10^{-7}
Cement 2	" " " " " "	1.58×10^{-6}
Cement 1	Pure Asphalt Cement 0.3 psi Pressure	2.31×10^{-6}
Cement 2	" " " " " "	2.76×10^{-6}

FIGURE 1. Effective Radon Diffusion Coefficient Versus Asphalt Content for Grand Junction Asphalt Cores

there is a fair amount of scatter, the diffusion coefficient was fairly constant over a range from 18.7 to 21.6%. This suggests that there is a relatively wide range of asphalts contents that could be used to obtain a successful radon seal. The one data point which did not fall into the same range as the others was the seal which contained 15.9% asphalt. This low asphalt content was due to the presence at a large chunk of clay contained in the center of the seal. This large chunk of clay raised the diffusion coefficient by a factor of 3 which stresses the need for quality control of the aggregate used along with close supervision of the front loader operators supplying aggregate to the cold mix paver.

The foamed asphalt core provided by FBDU was found to contain 29.7% asphalt. Even though this specimen contained ~50% more asphalt than the Grand Junction cores, its effective diffusion coefficient was a factor of 3 greater. One of the major factors in this difference is that it appears that a fine blow sand was used for this sample where as the field core samples used a well graded-1/4 in. concrete sand.

Earthen Cover Barriers

Material Characterization--M. E. Dodson and P. Reisenauer

A statistical analysis of the water content and bulk density for the multilayer plots was run. The soil parameters were the major ones that determine radon diffusion in soils. The results indicated that initial moisture content and the compaction achieved during the spreading and dumping operation are important factors in determining the variation in porosities and densities that can be observed in an earthen cover material. Measured characteristics indicated that radon diffusion in soil covers at normal field water contents, and packed to average density typical of undisturbed soils, will not be reduced by more than a factor of 10 to 50 below that determined for loose dry sand (approximately $5 \times 10^{-2} \text{ cm}^2/\text{sec}$).

Physical characteristics including particle size analysis and water retention properties were measured on 10 field samples supplied by Rogers and Associates. These samples were taken from soil profiles near Clive, Utah; Durango, Colorado; Riverton, Wyoming and Shiprock, New Mexico. Three other materials

taken from the earthen cover test plots at Grand Junction, Colorado were also characterized this quarter. These materials are potential candidate materials that may be used as earthen cover materials for tailings piles and are representative of a wide range of cover materials that are available near remedial action sites in the western United States. The data indicate that field water contents below 0.6 m are generally low except under shallow water table conditions. Air-filled porosities for all materials except at the Clive site were above 0.25. The Clive site has a shallow water table; hence, the clay soil was nearly saturated, with air porosities less than 0.10. These data will be useful in predicting long-term equilibrium moisture contents in cover materials at specific sites.

Radon Diffusion Tests--(RAE)

In order to compare radon diffusion coefficients measured by static or equilibrium methods with those measured by the transient technique, several intercomparison test columns were prepared using the same test soil. The soil used in the tests was Riverton sandy soil, compacted to a dry density of 1.62 g/cm^3 . One of the test columns was constructed previously with three ceramic cups and a hanging water column to control the moisture content despite evaporation from the exposed top surface of the soil. This column was monitored during the present quarter to obtain a total of six equilibrium radon flux measurements which corresponded to six different water column heights. The six radon diffusion coefficients calculated from these fluxes are plotted in Figure 2 for comparison with the calculated diffusion coefficients for this soil.

Two smaller-diameter test columns were constructed during the present quarter, and were equilibrated in a sealed configuration such that the radon passing through the soil was contained in the air chamber above it. After about six weeks, radon concentrations in the gas above and below the sample were sampled and used to compute diffusion coefficients which are plotted in Figure 2. The upper chambers of these columns were then removed, and the columns were allowed to re-equilibrate. Measurements of radon fluxes from these open systems were then used to compute additional diffusion coefficients for the same samples. A single ceramic cup in each of these small columns was used

FIGURE 2. Comparison of Measured Radon Diffusion Coefficients with Calculated Curves

to control the moisture by means of a hanging water column. The results of these measurements are also illustrated in Figure 2 and demonstrate reasonable agreement between the different diffusion measurements. Although the sealed-system measurements are slightly lower than the corresponding open-system measurement, the difference does not appear to be significant. Additional measurements are still in progress with the Riverton soil using the transient diffusion system.

Experimental data collected during the previous quarter on radon diffusion through water were also analyzed. These measurements utilized the sealed column approach for maximum sensitivity, with the water column being supported by a layer of agar containing a reinforcing screen. Three separate diffusion

coefficients were calculated for the three test columns to be 2.4×10^{-5} , 1.9×10^{-5} , and 2.0×10^{-5} when the distribution coefficient for radon between water and air was taken into account. The diffusion coefficients were 2.5-3 times lower if this distribution coefficient was ignored in the calculations. For comparison, a radon diffusion coefficient for water of $1.1 \times 10^{-5} \text{ cm}^2/\text{s}$ is reported in the literature.

Radon Diffusion Modeling--RAE and D. W. Mayer

Pore Distribution Modeling.

During the present quarter, a physicoempirical model recently reported by Arya and Paris (Soil Sci. Soc. Am. J. 45:1023-1030, 1981) was used to calculate pore size distributions from the sieve analysis data for eight soils where water retention curves were known.

Radon diffusion coefficients were calculated from the separate pore size distributions estimated from the drainage curve and sieve analysis data. These are plotted in Figure 3 for comparison of the diffusion coefficients. By analyzing eleven segments of each curve, it was noted that the overall standard error between the sieve analysis and water drainage curves amounted to a factor of 2.6, and that the overall bias amounted to only a factor of 1.18, with the sieve analysis curve being lowest. It is probably advisable to estimate diffusion coefficients from drainage curves rather than from sieve analyses whenever possible. This is due to the typically wider range of pore sizes generally estimated by the drainage curve data, and to the more extensive experimental verification of these estimates.

Defect Modeling.

Simulations of clay/gravel layer defects have been successfully completed for three configurations. The trace configurations consisted of 2, 8, and 16 randomly located defects occupying 2.5, 10, and 20% of the clay/gravel layer volume. Increases in surface radon flux due to defects ranged from a factor of 1.3 to 3 as the defects increased from 2 to 16. In comparison, when the entire area was defective the radon flux was increased to 6.5 times the value when no defect was present (Figure 4).

FIGURE 3. Comparison of Radon Diffusion Coefficients Calculated from Pore Size Distributions Based on Water Drainage Curves and Sieve Analyses for Shiprock, Riverton, Salt Lake City and Durango

2 DEFECTS

8 DEFECTS

16 DEFECTS

■ DEFECT REGIONS

FIGURE 4. Clay/Gravel Layer Defect Results

Model Verification.

Results from an analytical solution presented by RAE were compared with a two dimensional RADMD (PNL) solution. Input data for the model are shown in Table 4. Normalized flux and concentration values are shown in Figure 5. The surface flux values predicted by RADMD match those of the analytical solution with less than 1% error. Thus, RADMD has been verified for two dimension steady-state problems.

Coupled Flow Analysis.

The coupling of TRUST, a partially saturated flow code, and RADMD was completed. The first phase of the simulation (radon diffusion through a cross section of an uncovered tailings pile) has been completed and the response of the surface radon concentrations and flux to the placement of a cover has been determined. The results demonstrate model capabilities to assess both short term and long term effects of cover placed on tails which have been subjected to various stages of drainage.

FIELD TEST MONITORING

This quarters activity was mainly focused on planning and carrying out a field test monitoring trip to Grand Junction February 8-19. Work completed during the trip included 274 radon flux measurements, neutron probe moisture measurements on the FY80 and FY81 test plots, 2 complete sets of radon concentration measurements in the columns, data logger maintenance, and manual reading of moisture blocks and salinity sensors not hooked up to the data loggers.

Other related activities this quarter include development of data analysis and plotting programs and analysis of selected temperature, capillary pressure, and water content profiles accumulated to date.

Radon Flux Measurements--H. D. Freeman and R. R. Kirkham

Radon flux measurements were made on the FY80 Multilayer and Asphalt covers, and FY81 Joint Field test areas (260 total measurements). A summary of the radon flux measurements is shown in Table 5 and Figure 6. The measurements were made on the radon barriers using the PNL flow through systems, with a 4 hr

TABLE 4. Layer Characteristics

Layer	Thickness, cm	Porosity	Moisture Content, cm ³ /cm ³	De, cm ² /s
Sand	15	0.35	0.00	0.0123
Tailings	45	0.48	0.05	0.0121

Layer	D, cm/s ²	Radium Content, pCi/g	Emanating Power	Bulk Density, g/cm ³
Sand	0.0350	0.0	0.2	1.755
Tailings	0.0279	1092	0.2	1.400

FIGURE 5. Surface Flux and Concentration Distribution

TABLE 5. Radon Flux Values for FY80 and FY81 Radon Barriers

Barrier	pCi/m ² .s	
	Flux Range	Avg $\pm$ SD
FY81 Multilayer		
15 cm Gel Mix	1.09-29.3	12.0 $\pm$ 8.05
30 cm Gel Mix	17.9-55.4	32.2 $\pm$ 10.9
40 cm Bentonite/Lime Mix	8.84-35.5	19.2 $\pm$ 7.99
30 cm Bentonite Mix	12.5-66.6	38.2 $\pm$ 17.7
FY81 Asphalt Emulsion	0.16-183	9.88 $\pm$ 28.5
FY81 Earthen Covers		
Mancos Shale	0.54-3.46	1.38 $\pm$ 0.95
Bentonite	1.86-22.8	11.1 $\pm$ 6.30
Compacted Adobe	1.80-32.61	10.6 $\pm$ 10.0
Uncompacted Adobe	2.78-53.7	24.1 $\pm$ 17.0
FY80 Multilayer		
Gravel Moisture Barrier	0.23-0.79	0.42 $\pm$ 0.16
No Gravel Moisture Barrier	2.22-109	32.0 $\pm$ 33.0
FY80 Asphalt Emulsion	0.05-12.02	1.04 $\pm$ 2.68

sampling period. Approximately 5 cm of snow was swept from an area around each of the FY81 test points to expose dry soil for the measurement. The measurements on the 1980 test plots were made on wet soil. The activated carbon samples from the measurements were shipped to PNL for counting on the intrinsic Germanium diode.

Neutron Probe Measurements--R. R. Kirkham and H. D. Freeman

All neutron moisture meter access wells were brought to a constant height of eight cm above the soil surface. This will simplify the positioning of the neutron moisture probe in each access well. A new Troxler moisture probe was cross-calibrated with the existing Campbell Pacific Nuclear probe. To improve calibration data for the neutron probes, one water standard with interchangeable aluminum and steel pipes, and two dry soil barrels, one each with aluminum and steel pipes, were constructed. Measurements were made in these barrel

FIGURE 6. Radon Flux Values for 1981 Joint Field Test, February 1982

standards and will be used to develop a moisture calibration for aluminum and steel access pipes used at Grand Junction. Bimonthly readings will be made by Bendix. These began in March.

Data analysis from neutron probe logging of field columns earthen covers and multilayers continued. Typical results of these logs in the earthen cover field plots and in the columns are shown in Figures 7 and 8. The data indicate that some drying of the compacted wet layers are occurring in the columns. No significant drying with depth seems to be occurring in the field plots, however, additional data is needed to verify this. The columns appear to initially have higher water contents in the compacted zones than those for similar test in the field plots. On the other hand there is a higher water content in the Adobe cover in the field plot than in the column tests making it difficult to directly compare the radon flux data from the columns and field plots.

Temperature.

Typical temperature profiles in earthen covers are shown in Figure 9. The data indicate that during the monitoring period (December through February) the frost line (0°C isotherm) did not move below 0.6 m (2 ft) depth in any of the earthen cover. Column data compared well with the data obtained from adjacent field plots. Little difference in thermal profiles existed between different cover treatments. This is likely true because of the similar nature of the cover material (Adobe clay).

Capillary Pressure.

Typical capillary pressure curves are shown in Figure 10 for the earth cover column tests. The capillary pressure (measured with electrical resistance gypsums blocks) indicate the state of soil dryness. The higher pressure values in the uncompacted adobe reflect the dry nature of the material over the entire cover depth. Increased wetness (lower pressure) is indicated in the compacted layers of the other cover systems. The capillary pressure, temperature and water content in these covers will continue to be recorded with time to determine the effect of the climatic parameters on earthen cover systems and help assess the changes that may occur in surface radon flux.

FIGURE 8. Water Content Profiles in Earthen Covers (Compacted Adobe)

FIGURE 9. Temperature Profiles in Earthen Covers (Column Tests)

FIGURE 10. Capillary Pressure Profiles in Earthen Covers (Column Tests)

Column Monitoring--H. D. Freeman

Column monitoring included radon flux measurements, radon concentration profile measurements, neutron probe measurements and servicing of the data logger.

Two radon flux measurements were taken on each column the results of these measurements are shown in Table 6.

Soil radon concentration measurements were made twice in each column. Results of these measurements are presented in Figure 11. These concentration profiles will be used to verify radon concentration modeling now underway.

The data logger located in the access column had ceased to function in mid-January. This problem appears to be some type of intermittent fault which prohibits proper operation and manual restart commands. The unit was returned to service, but the source of failure was not determined. If during biweekly servicing of the unit by Bendix staff, failure is again detected, the unit will need to be returned to the manufacturer for servicing.

TABLE 6. Radon Flux Measurements for the 7 Column Tests

Column		Radon Flux		
		pCi/m ² s		
		1	2	Avg.
A1	Asphalt	2.18	0.72	1.45
M1	30 cm Alum Mix	5.83	11.4	8.61
M2	30 cm Bentonite/lime Mix	10.6	7.87	9.23
EC1	Mancos Shale	22.3	14.0	18.1
EC2	Bentonite	25.0	21.5	23.2
EC3	Compacted Adobe	40.1	46.4	43.2
EC4	Uncompated Adobe	14.9	20.3	17.6

FIGURE 11. Column Radon Concentration Profiles February, 1982

ENGINEERING SPECIFICATIONS

Cost Estimates--E. G. Baker

The purpose of this task is to compare the cost of different radon barrier systems and determine where cost reductions can be made. In this past quarter we compiled the 1981 Grand Junction field test costs for the earthen, asphalt, and multilayer covers systems being tested. We also completed cost estimates of earthen and asphalt covers for the Durango, Shiprock, and Vitro sites. We will calculate the cost of a multilayer cover at the three sites during the next quarter. A draft of a report on the cost of radon barrier systems should be complete by the end of next quarter.

The actual costs incurred during the 1981 Grand Junction field test are shown in Table 7. These are the latest, most complete costs and are slightly revised from those presented in the last monthly report. These costs include all subcontractor costs, materials, equipment, and PNL labor costs incurred at the site. PNL costs associated with engineering and planning the field test are not included. For the earthen and multilayer systems, these costs are an average of the four different systems tested. The range of costs for the different systems is also shown. The mancos shale and adobe clay were the least expensive earthen covers. A 15 cm gel system was the least expensive multilayer cover.

TABLE 7. Grand Junction Field Test Costs

	<u>Units</u>	<u>Earthen Covers</u>	<u>Asphalt Cover</u>	<u>Multilayer Cover</u>
Site Preparation and Maintenance	\$/m ²	2.50	3.20	2.56
Cover Application	\$/m ²	7.70	7.20	22.30
Materials	\$/m ²	16.70	16.80	32.30
Total Cost	\$/m ²	26.90	27.20	57.10
Range	\$/m ²	(25.5-32.8)	--	(42.8-64.8)
Area Covered	m ²	2500	4900	3250
Total Cost	\$	67,200	133,200	185,600

Cost estimates for a 3 m earthen and a 7.5 cm asphalt cover (including 0.6 m burden) systems at Durango, Shiprock, and Vitro are presented in Table 8. We selected two disposal locations for each site based on engineering assessments completed by Ford, Bacon, and Davis Utah, Inc. Care must be exercised in the use of these site specific cost estimates because insufficient data and information are available at this time to completely characterize the site. These estimates do provide a general idea of what the radon barrier systems will cost and provide a means to compare the various cover systems.

The costs in Table 8 include a contingency and engineering and contracting expenses. They are less than the cost of the Grand Junction field test because of more efficient utilization of labor, equipment, and materials. The earthen cover costs are also less because of the assumption that acceptable cover material is available closer to all the sites and thus cheaper than the cover material that was used at Grand Junction.

Materials and Application Specifications--E. G. Baker

The major objective of this task is to develop engineering specifications ready for use by a remedial action contractor. The emphasis this quarter has been on the development of laboratory mix design procedures to evaluate the setting characteristics of various combinations of emulsions and aggregates.

TABLE 8. Site Specific Cost Estimates for Earthen and Asphalt Radon Barrier Systems

Site	\$ / m ²	
	3 m Earthen Cover	7.5 cm Asphalt Cover
<u>Durango</u>		
East Ridges Basin	18.20	25.30
Bodo Canyon	20.40	24.50
<u>Shiprock</u>		
Many Devils	18.40	24.60
Rattlesnake	22.00	25.70
<u>Vitro</u>		
Clive	16.10	24.00
Sager's Flat	23.60	23.80

A mix design test apparatus has been developed and is shown in Figure 12. It consists of a Servodyne drive system, which includes a DC motor-generator and a solid state electronic controller, and a containment vessel. The servodyne controller maintains the stirrer at a constant speed and provides a read-out of the torque. The aggregate and emulsion are mixed in the vessel and as the admix begins to set the torque increases. Each formulation of aggregate and emulsion will have its own characteristic "setting curve".

A mixer configuration was developed which mixes the dry aggregate well without stalling the mixer and still provides sufficient sensitivity to measure the setting properties. Various formulations will now be tested to define their setting characteristics and to determine how the setting time can be controlled. The majority of this work is being performed by Petroleum Sciences, Inc. However, a similar system as been set up at PNL to verify the repeatability of the test.

FIGURE 12. Laboratory Mix Design Test Apparatus

QUARTERLY REPORT
LINER EVALUATION FOR URANIUM MILL TAILINGS

J. L. Buelt - Project Manager

The objective of the liner program is to evaluate prospective materials for preventing moisture transport of radionuclides and hazardous chemicals from uranium mill tailings over a long-term period. The approach embodies liner criteria development and material performance evaluations through laboratory and field studies.

SUMMARY

Recent efforts have concentrated on identifying the contaminant source terms from the tailings and quantifying the retardation characteristics of clays and soils. A comparative cost estimate has been tabulated for various types of liner materials. This information is necessary input for the decision tree being developed to identify the need and type of liner at a disposal site.

LONG-TERM PERFORMANCE TESTING - J. F. Relyea, W. J. Martin and D. T. Colclough

A. Tailings Characterization

Three tailings samples have been characterized using a column leaching method. Most of the heavy metal and radioactive contaminants were released in the first pore volume of leachate from tailings columns that simulate inactive tailings piles. Concentrations of As, Ba, Cr, Pb and NO₃ from the acidic Shiprock and Salt Lake tailings shown in Table 1 were below the maximum permissible concentration levels (MPC) for groundwater at the site boundary, (proposed by EPA in 40 CFR Part 192, January 9, 1981), after only two pore volumes of leachate had been collected. Cadmium concentrations were still a factor of ten higher than the MPC. Leachate concentrations of Ba, Cd, Cr, Pb, and NO₃ from the alkaline Durango tailings, shown in Table 1, were all below MPC values after two pore volumes but arsenic remained above its MPC (the elements Hg, Se and Ag were not analyzed).

Radioactive element concentrations from acidic tailings were also found to drop rapidly in effluent from the columns, but ²³⁸U and ²³⁵U concentrations in effluent from the alkaline Durango tailings remained fairly constant out to 1.4 pore volumes.

Detection limits for the radioactive elements were higher than MPC values because of the relatively small sample volumes. To properly assess the contaminant inventory of a tailings sample, small columns requiring about 300 to 500 g of tailings can be used for the non-radioactive elements. Larger columns holding 2 to 3 kilograms of tailings must be used to obtain sufficient leachate samples for detection of radioactive elements near or below their MPC values.

We may conclude that the contaminant release rate and inventory of a tailings pile may be estimated using the column leach method. Samples of the tailings from a site should be taken at several locations and depths to characterize the variability of the tailings. These data may be used to formulate a "worst case" contaminant release for input to the decision tree.

B. Liner/Soils Characterization

Two soil samples from Clive, Utah have been characterized. The most outstanding characteristic of both soils is their high soluble salt content which indicates the upward movement of groundwater to the surface. Soluble salts were determined using a 1 to 1 soil to water ratio extraction method (Bower and Wilcox, 1965). From 0.5 to 1.0 grams of soluble salts were found per 100 g of soil. Sodium accounted for over 80% of the soluble salts which could cause the soil to disperse if contacted with water. This information is important because the saturated hydraulic conductivity of the soil may be reduced significantly if it is dispersed on contact with water from a heavy rain or low ionic strength groundwater. The behavior of the soil may however be different if contacted with high ionic strength leachate from an inactive tailings pile. If these soils are to be considered as liner or cover materials, their hydraulic conductivities must be studied with both low ionic strength water and higher ionic strength tailings leachate so that both infiltration of rainwater or groundwater and leachate movement can be predicted with hydrologic modeling techniques.

The buffering capacities of these soils were measured by HCl titration of one gram samples in fifty milliliters of distilled water. From five hundred to more than seven hundred meq of HCl were required to reduce the soil suspension pH to 2.0.

C. Leachate-Soil/Liner Interactions

The first 5:1 batch leachate-soil interaction tests have been sampled but solution analysis is incomplete. Values for pH predicted from the soil buffering capacities are lower than pH values measured in the acidic leachate-soil suspensions. This indicates the actual soil buffering capacities for acidic leachate are greater than the buffering capacities for HCl. The HCl buffering capacity could be used in a conservative (worst case) prediction of the migration of acidic leachate from the tailings pile, however, the actual soil buffering capacity for leachate may yield a more realistic prediction.

Effluent has been collected from most of the soil/liner permeability tests. Values for pH in the initial samples varied from 7.4 to 8.2 but may not be too reliable because of sodium interference from the high salt concentrations. Electrical conductivities varied from 60 to more than 100 mmhos/cm which corresponds to an ionic strength of near one molar NaCl.

Saturated flow through the column is apparently concentrating the soluble salts from the soils and liner material in the advancing water front. The flow rates are also decreasing in several of the columns, which could be caused by either: 1) dispersion of the sodium saturated soil, or 2) plugging of the pores near the leachate input port by precipitation of solids as the leachate pH increases. It is not clear whether or not this decrease in permeability would be observed at the Clive site, because water movement may be by upward unsaturated rather than saturated downward flow. This points out the need for well planned site characterization in the Decision Tree procedure discussed below.

In summary, the following conclusions must be regarded when designing an impoundment system for inactive mill tailings.

- Concentration levels for most primary drinking water contaminants (10 CFR 143) are below the MPC levels after only two pore volumes of leachate have been collected from tailings columns.
- Radioactive contaminant concentrations from acidic tailings columns drop off rapidly as the tailings are leached.
- The contaminant release rate (concentration vs pore volumes of leachate eluted) and inventory (total contaminants released) from a tailings sample are limited and can be estimated using the column leaching method.

- Large columns (2-3 kg of tailings) are required for characterizing the release rates of radioactive elements. Large columns are needed to give the large sample volumes (200-300 ml) required to lower detection limits to the MPC values proposed by EPA.
- Actual buffering capacities of soils for tailings leachate are larger than the buffering capacity measured by acid titration.
- Site characterization must be an iterative process so that limited resources can be expended gathering the most important data for predicting water and contaminant transport. This helps to eliminate gathering expensive data that is later found to be unnecessary.

LINER CRITERIA DEVELOPMENT (DECISION TREE) - J. F. Relyea and S. E. Petty

A draft decision tree was presented last quarter and has undergone an internal peer review. It has become clear that the site characterization plan must be an iterative plan.

The objectives of the site characterization plan is to gather data needed to predict possible water movement into the tailings and subsequent movement of contaminants with tailings leachate. The first step in this process should be to gather all available data on a proposed disposal site so that a conceptual water transport model can be delineated. At the Clive site for example, saturated flow may dominate thirty feet below the surface, but the net water flux may be upward unsaturated flow to the surface where salts are deposited by evaporation. The data needed for this flow system may be very different than that needed if we had assumed water would come only from rainfall. Once the conceptual model is described, a cost effective site characterization plan may be developed to gather pertinent data for the site hydrologic model (in other words, data that will not be needed does not have to be measured).

COST COMPARISON - S. M. Barnes

A preliminary installed cost comparison has been completed for several liner materials at a site in Clive, Utah. The cost comparison is presented in Table 2 and should be used for comparison, only. The data was collected from various manufacturers and installation contractors. The analysis was based on the following assumptions:

- A 210,000 m² tailins impoundment at Clive, Utah.
- Soil suitable for mixing with the sodium bentonite to form the clay liner was available on the impoundment site at no cost.
- Mill tailings would be installed above the polymeric and asphalt liners for protection against heavy equipment damage.
- The site preparation cost is constant for each of the liners.

TABLE 2: Installation Costs for Various Liner Materials at Clive, UT Site

<u>Material</u>	<u>Application Rate</u>	<u>Cost, \$m²</u>
Chlorosulfonated Polyethylene (Hypalon)	30 mil	5.92
High Density Polyethylene	30 mil	4.63
Catalytic Airblown Asphalt	9 kg/m^2	2.58
Gilsabind Membrane #6, Rubberized Asphalt	100 mil	4.31
Sodium Bentonite Clay	19.6 kg/m^2	2.26

REFERENCES

Bower, C. A. and L. V. Wilcox. 1965. Soluble Salts in "Methods of Soil Analysis," Part 2, pp. 933-951. C. A. Black, Ed. American Society of Agronomy, Madison, Wisconsin.

TABLE 1. Comparison of Contaminant Concentrations
in Tailings Leachate with EPA Groundwater
Criteria

	Acidic Tailings								Alkaline Tailings				Detection Limits (ppm)	MPC(a) (ppm)
	Salt Lake				Shiprock				Durango					
Pore Volume	0-0.2	0.2-0.4	0.9-1.1	2.2-2.4	0-0.3	0.3-0.5	0.9-1.1	1.7-1.9	0-0.2	0.2-0.4	1.2-1.3	1.8-2.0		
pH	2.0	2.1	2.4	2.6	2.8	2.5	2.9	3.0	9.5	9.7	9.0	8.8		
TDS (g/liter)	18	7.2	4.3	3.4	42	12	3.8	2.6	4.6	1.9	0.4	0.3		
As (ppm)	12	1.4	nd	nd	11	2.6	nd	nd	16	10	1.4	0.4	0.08	0.05
Ba (ppm)	nd	nd	nd	nd	nd	0.1	nd	0.1	0.03	0.03	0.03	0.03	.002	1.00
Cd (ppm)	2.6	1.0	0.3	0.1	5.3	1.5	0.2	0.3(b)	0.15	0.10	0.01	nd	.004	0.01
Cr (ppm)	2.9	0.6	nd	nd	1.9	nd	nd	nd	1.6	0.2	0.02	nd	.02	0.05
Pb (ppm)	nd	nd	nd	nd	nd	1.7	0.1	nd	nd	nd	nd	nd	.06	0.05
NO ₃	nd	nd	nd	nd	690	170	nd	nd	7	nd	nd	nd	1.0	10.00
(a) MPC = Maximum permissible concentration for groundwater at the site boundary as established by EPA in 10 CFR 192.														
(b) Cd detected in effluent from pore volume fraction from 2.5 to 2.8.														

REVEGETATION/ROCK COVER FOR STABILIZATION OF INACTIVE U-TAILINGS SITES

P. A. Beedlow - Project Manager

The objective of this program is to establish guidelines for surface stabilization that provides for compatibility with sealant/barrier systems and is suited to soils and climates at inactive uranium tailings sites.

SUMMARY

Root distributions were sampled on the 1980 test plot at Grand Junction. Roots were present at all depths in the overburden and had penetrated into the surface of the tailings in areas with no radon seal. Six inches of rock was applied to the 1980 multilayer earth plot to study the effects of rock cover on soil moisture. Observations made in early October of the San Juan test plot indicate that two to four times as many plants died on the rock cover treatment as on bare soil. In early March, a meteorological station and data logging system were installed on the San Juan test plot and hydroprobe readings were taken. The field sampling work of the regional evaluations task was completed in March. Analysis of the soil samples and vegetative cover measurements were completed. The data have been summarized and are now being used to group the sites into regions and to construct a model of revegetation potential.

TEST PLOTS - P.A. Beedlow

Interactions between surface treatments and sealant/barrier systems are being studied at the Grand Junction Test Site in conjunction with UMTRA projects testing sealants and biological barriers.

Root distribution studies were conducted at the Grand Junction Test Plot in late October. Trenches were dug on the asphalt portion of the plot. Four trenches were located in three to four feet of overburden (two over asphalt and two just off the edge), and three trenches were in one to two feet of overburden (two over asphalt and one over tailings). Root distributions were estimated by sampling root frequency at 20 cm depth increments. Gravimetric soil moisture and bulk density samples were taken at the same locations. Roots

were present at all depths in the overburden and had penetrated into the surface of the tailings in areas without an asphalt seal. No damage to the asphalt seal was apparent. Soil cores were weighed, dried and reweighed for gravimetric soil moisture and bulk density measurements. These data are currently being analyzed to determine if soil moisture, soil density, and depth of overburden significantly affect rooting patterns.

In conjunction with the multilayer barrier program for sealing uranium tailings, a rock cover was added to a portion of the 1980 test plot at Grand Junction. A layer of 2 in. washed rock was applied 6 in. deep over half of the plot. Measurements of soil moisture will be made with soil moisture blocks and a neutron probe. This work will provide moisture profile data for validation of the unsaturated flow model (UNSATV). This model is being used to predict the water accumulation in and drainage through cover/tailings as a function of rock and vegetation surface stabilization treatments.

The effects of irrigation and rock covers on soil moisture and plant establishment are being evaluated at a second test site on the San Juan coal mine near Farmington, N.M. In early October a visit was made to the San Juan test plot to observe conditions following the fall irrigation period. This test site was generally in good condition. Numerous rills were noted on the south and north sides, a result of the general slope of the plots. On plots that had been covered with rock, the rills were filled with rocks that had settled into the erosion furrows, thus restricting further erosion. For 648 transplants, mortality was as follows:

<u>Chrysothamnus nauseosus</u>	80 = 12.5%
<u>Rhus trilobata</u>	30 = 4.6%
<u>Juniperus scopulorum</u>	15 = 2.3%
<u>Atriplex canescens</u>	18 = 2.8%

Initial observations indicate that two to four times as many plants died on the rock cover treatments as on the bare soil. Confirmation of actual survival percentages must be delayed until spring because some species will resprout from the base. *Rhus trilobata* transplants often appear dead because leaves drop off when the plants are stressed.

In early March a meteorological station and data logging system were installed on the San Juan test plot. In addition hydroprobe readings were taken and portions of the test plot damaged by water erosion were repaired.

The data logging system has the capability to continuously monitor air temperature, relative humidity, solar radiation, wind speed, wind direction, rainfall, soil moisture, and soil temperature.

Eighteen gypsum soil moisture blocks were located in two treatment plots (bare soil and rock mulch). Three replicate blocks were located in 3 areas receiving high and low amounts of irrigation water. Blocks were placed in the top 5 inches of the soil, 2 inches above and 2 inches below the soil-spoil interface. Thermistors were placed in the center of the two treatment plots. Placement depths were: surface, 5, 14 and 20 inches.

The data obtained from these instruments will be used to validate the unsaturated flow model we are using to estimate soil moisture dynamics under rock and vegetative covers.

DATA ACQUISITION AND REGIONAL SAMPLING - M.C. McShane

Regional sampling and analysis of soils and vegetation in the vicinity of the inactive sites is being conducted to determine the effects of soils and climate on vegetative cover. These data will be used to determine the environmental factors controlling vegetative cover. From this information a model for estimating revegetation potential will be constructed. Determining potential vegetative cover in areas of the inactive sites is necessary for estimating the nature and extent of erosion. With this knowledge, surface treatments can be designed to minimize erosion and contribute to effective containment of toxic components.

The bulk of the field sampling was completed summer, FY-81 and the remaining region, the south Texas Coastal plain was sampled in late March, 1982. During the first half of FY-82, most major steps in the regional data analysis were completed:

- The results of the chemical and mechanical analysis of all the soil samples were received from the subcontracted soils laboratory. These included pH values for 400 surface samples, nutrient and textural analysis of approximately 600 samples and particle size determination of about 360 samples.
- Computer programs needed to summarize the soils data and analyze the vegetative cover measurements were written and are in use.

- A cluster program was obtained from Utah State University and modified for application to the site climate, soils, and vegetative cover data.
- Sites were clustered into climatic regions and grouped according to vegetative type and cover.
- Canonical analysis (or ordination procedure) of all the collected data was initiated to determine specific environmental factors controlling vegetative cover and type.
- Work was begun on correlation and variable reduction procedures which will ultimately be used to construct a model for estimating vegetative cover and revegetation potential.

APPLICATION OF LONG-TERM CHEMICAL
BIOBARRIERS FOR URANIUM TAILINGS

J. F. Cline - Project Manager

The objective of this project is to develop and evaluate the effectiveness of physical and chemical barriers that are designed to prevent plant and animal breachment of uranium tailings containment systems.

SUMMARY

Release rate of trifluralin from the PCD pellets used in the field tests at Grand Junction U-tailings was measured in the continuous flow system and in the soil placed in lysimeters. The release rates were affected by temperature. Also, a burst of trifluralin was released from the pellets at the beginning of the test due to quantities of trifluralin adsorbed to the surface of the pellet. After the initial burst the release rates were continuous and reacted as calculated.

Root elongation was stopped in the soil over the pellet layers containing pellets placed on one and two inch centers in a plant growth chamber study.

DETERMINE HERBICIDE RELEASE RATES FROM POLYMERIC CARRIER/DELIVERY SYSTEMS (PCD)

The release rates of trifluralin from the PCD system placed over the containment systems in August 1981 at the Grand Junction U-tailings were determined using the continuous flow system. The release rates measured immediately after the pellets' fabrication were as follows:

<u>Temperature (°C)</u>	<u>Release Rate ($\mu\text{g/day} \pm \text{SD}$)</u>
37	853 $\pm$ 91
22	131 $\pm$ 8
13	29 $\pm$ 2

Following these initial analyses, the pellets were held at 37°C in the continuous flow system for 54 days. During the last 18 of the 54 days, the release

rate (at 37°C) was 198 ± 32 , as expected, since the trifluralin must now diffuse for some distance within the device, leading to a lower release rate. Following this 54 day run at 37°C, the temperature of the bath was lowered to 13°C, the nominal temperature at 1 to 2 meters below ground level in arid regions. Sufficient trifluralin had been released at this point to simulate a device which had been releasing trifluralin for five years. The determined release rate (13°C) at this point was 13.1 ± 2.6 .

The release rates of trifluralin and the effectiveness as a root barrier of the fabricated pellets used at the Grand Junction site were tested in soil placed in lysimeters. The pellets were placed in small lysimeters (15 cm in diameter and 50 cm long) in a layer in the soil 40 cm below the soil surface at the same rates (on 1 inch and 2 inch centers) as used in the field studies. The lysimeters were then planted to oats, placed in a plant growth chamber and allowed to grow for 90 days. The distance was measured between the pellet layer and the point where the root elongation stopped above the pellet layer. Root elongation stopped 3.5 cm and 1.6 cm above the layers where pellets were placed on one and on two inch centers, respectively. This verified in a controlled experiment that pellets placed on two inch centers would stop root elongation and prevent root intrusions of the seals placed over U-tailings. These observations will be compared at a later date to observations made of pellets placed in the overburden above the asphalt emulsion and multi-layer soil seals installed over the Grand Junction U-tailings in August 1980.

FINAL REPORT

The rough draft of the final report is nearly complete. Chapter I through IV have been given to the editor for review and preparation for publication.

PUBLICATIONS

The document entitled "Habitat Requirements and Burrowing Depths of Rodents in Relation to Shallow Waste Burial" by K.A. Gano and J.B. States is being submitted for clearance under document number PNL-4140.

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