A Decision Tree Approach to Evaluating Inactive Uranium Processing Sites for Liner Requirements

J. F. Relyea

March 1983

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Pacific Northwest Laboratory
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ABSTRACT

Recently, concern has been expressed about potential toxic effects of both radon emission and release of toxic elements in leachate from inactive uranium mill tailings piles. Remedial action may be required to meet disposal standards set by the states and the U.S. Environmental Protection Agency (EPA). In some cases, a possible disposal option is the exhumation and reburial (either on site or at a new location) of tailings and reliance on engineered barriers to satisfy the objectives established for remedial actions. Liners under disposal pits are the major engineered barrier for preventing contaminant release to ground and surface water. The purpose of this report is to provide a logical sequence of action, in the form of a decision tree, which could be followed to show whether a selected tailings disposal design meets the objectives for subsurface contaminant release without a liner. This information can be used to determine the need and type of liner for sites exhibiting a potential groundwater problem.

The decision tree is based on the capability of hydrologic and mass transport models to predict the movement of water and contaminants with time. The types of modeling capabilities and data needed for those models are described, and the steps required to predict water and contaminant movement are discussed. A demonstration of the decision tree procedure is given to aid the reader in evaluating the need for and adequacy of a liner.

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1.0 INTRODUCTION

Disposal criteria have recently been drafted to protect the public from potentially harmful effects of radon emissions and release of toxic elements in leachate from inactive uranium mill tailings piles. Remedial action at several inactive tailings sites may be required to reduce potential adverse health effects to acceptable levels determined by state agencies and the EPA. Several disposal options involve exhumation and burial of the tailings (either on site or at a new site) using various engineered barriers including liners. The barriers are designed to prevent water and/or contaminants from moving into or out of the tailings pile, thus limiting the release of contaminants from tailings to ground or surface water. This is accomplished by both reducing the flow and chemically attenuating (i.e. adsorption and/or precipitation) potential contaminants.

The purpose of this report is to provide a logical sequence which will help determine whether a liner is required for a specific site and to facilitate design specifics to meet the regulatory criteria for subsurface contaminant release levels. Potential users of this document may come from a wide variety of backgrounds ranging from architect and engineering (A&E) firms to legal and regulatory personnel. Design firms can use the document as a tool or framework about which they can logically arrive at the proper design for a specific site. Regulatory personnel may use the document as a tool for ensuring that a proper approach has been used by design firms and that site development by construction firms reflects the proper design. The intent of this document is to outline an approach which will be comprehensive enough to protect the environment near all disposal sites with groundwater contamination potential, and yet still be flexible enough to account for site-specific details.

Uranium production in the United States has traditionally led to the generation of large volumes of spent ore known as uranium mill tailings. During the period from 1940 to 1960, uranium surface mining facilities were primarily located in the western states of Colorado, Utah, New Mexico and Wyoming. Many of these mining and milling facilities are now inactive. Because the uranium ore contained relatively small percentages of uranium, the milling of large

quantities of ore was required to extract a small amount of product, commonly called yellowcake. In fact, only about 0.15% of the ore was converted to yellowcake. The remaining 99.85% of the ore mined became waste material or "tailings" as it is often called.

Over 25 million tons of tailings have been produced during the past 40 years. These tailings were usually disposed of by dewatering and stacking in piles adjacent to the ore refining facilities. Once production was discontinued, either because of ore depletion or low demand, the tailings piles were simply abandoned.

Release of heavy metals and radioactive elements to surface and ground waters may be found to be a problem at a few mill tailings sites and may need to be reduced in order to protect such waters for public usage. The increased surface area of the crushed ore, the altered chemical state of the spent ore and associated fluids generated during the highly acidic uranium leaching process, the concentration of the tailings piles within or above potentially potable water systems and the effects of mechanical, chemical and biological weathering of spent ore piles all contribute to the potential environmental hazards via surface and ground-water transport paths.

In 1978, because of the potential radiological and chemical hazards posed by inactive uranium mill tailings piles, the U.S. Congress passed legislation directing the Department of Energy to enact a plan which would secure uranium mill tailings piles in an environmentally acceptable manner.

The goal of this paper is to present a methodology through which inactive uranium mill tailings sites can be evaluated. The methodology might be used where potential beneficial use of the groundwater exists and where a preliminary assessment suggests the possibility of groundwater contamination from the tailings. This methodology will reflect the fact that several different techniques may be used to prevent or mitigate releases from uranium mill tailings sites to satisfy criteria proposed by state or federal regulatory bodies. The methodology proposed for site evaluation sets the stage for developing recommendations that are most practical from cost, technical and regulatory standpoints (regardless of the final form of the disposal regulations).

2.0 DESCRIPTION OF AN APPROACH TO SITE EVALUATION FOR LINER REQUIREMENTS

To evaluate a disposal site for liner requirements, an understanding must be developed of the processes that can potentially remove contaminants from the tailings and transport those contaminants to surface or ground water. These processes are outlined in Figure 1.

If water infiltrates the tailings pile, the soluble contaminants will be leached out and move with the water. Contaminant concentrations in the leachate are controlled by the quality of water entering the tailings, the types of minerals in the ore and the processes used to extract uranium from the ore. Subsequent contact between tailings leachate and liner material or sub-soil may then lower contaminant concentrations by several mechanisms. Precipitation of contaminants is especially important for low pH (acidic) leachate.

Neutralization of acidic leachate on contact with soils or liner material having a neutral or alkaline pH can reduce contaminant concentrations by one or more orders of magnitude. These reactions are controlled by the pH, ionic strength, chemical composition of the leachate and the buffering capacity of the soil.

Contaminant concentrations may also be reduced by adsorption of contaminants on liner and soil surfaces. This mechanism is more important for neutralized leachate and for leachate from alkaline processed ore. Adsorption is generally controlled by the surface area of the liner material or soil, the solution pH and ionic strength and concentrations of competing and complexing ions.

Reductions in contaminant concentrations by dilution may be observed if leachate contacts moving ground water. Moving ground water may further reduce these concentrations at the contaminant front by hydrodynamic dispersion.

The purpose of this section is to describe a decision tree approach that can be used to predict the movement of contaminants based on the processes mentioned above. The predictions of contaminant migration can then be used for evaluation of a site for liner requirements.

FIGURE 1. Important Processes that Control Release of Contaminants from Uranium Mill Tailings to Ground and Surface Water

2.1 HYDROLOGIC MODELS

Where a potential groundwater problem exists, methods are needed that predict whether particular disposal site designs are adequate to mitigate leachate migration past fixed boundaries during long-term storage. One method that can be used to predict the movement of moisture and contaminants with time is the use of computer models. Modeling of both hydrology and contaminant transport

is done quite frequently. Both saturated and unsaturated flow hydrologic models are available. Simmons and Gee (UNSATV) (1981); Bond, Freshley and Gee (UNSAT1D) (1982); and Reisenauer et al. (TRUST) (1982) describe in detail various hydrologic models that can be used to predict unsaturated flow through soils. The saturated flow model MMT is described by Ahlstrom et al. (1977).

In many cases, water movement through a tailings disposal site, particularly in the arid western United States, may be insignificant. However, where waste water movement exists, it is likely to occur by unsaturated flow. For a site where models may be useful, a good approach may be to combine an appropriate unsaturated flow model to predict the effect of climate, surface layers and vegetation on infiltration to the tailings with another model. This model would use the infiltration as a boundary condition for prediction of water movement through the tailings and subsurface layers. The model used to predict subsurface flow would depend on conditions at a specific site (e.g., saturated or unsaturated water movement).

2.2 CONTAMINANT SOURCE TERM

To decide whether a liner will be needed at a particular site using decision tree methodology, the quantities and release rates of contaminants from the tailings must be determined. Without the data that describe the contaminant source term, no evaluation can be made of contaminant concentrations entering subsurface or surface waters.

Contaminant leach rates have been studied by flushing water through columns packed with tailings samples from several locations (Relyea, Martin and Colclough 1982). Generally, contamination concentrations are highest in the initial pore volumes of leachate and decrease to much lower levels after one or two pore volumes of tailings leachate have been generated. Contaminant release rate and total contamination released from a tailings pile are often estimated using this column leaching method.

2.3 CONTAMINANT ATTENUATION

The movement of contaminants from a tailings disposal site to subsurface or surface water can be predicted by simply assuming that the contaminants move

at the same velocity as the water released from the tailings. This is a worst case scenario which ignores the fact that many chemical reactions between tailings leachate and soil can greatly reduce contaminant concentrations. The most common technique for predicting contaminant migration is to use a retardation factor. A retardation factor is the most simplistic way to predict the relative migration velocity of a contaminant to the velocity of water. For example, a retardation factor of 100 implies that water travels 100 times faster than the contaminant in question. The retardation factor approach is strictly valid only when several conditions are met: no precipitation occurs; adsorption is reversible and can be described by a linear adsorption isotherm; chemical composition of the solution is constant in both time and space.

For the many cases in which acidic tailings leachate may contact neutral or alkaline soils, the conditions under which a retardation factor is valid are not met. However, studies of leachate-soil interactions have shown that most contaminant concentrations are greatly reduced when the leachate pH is raised on contact with soil (Gee et al. 1980). By using measured values for the soils buffering capacity and total acidity in tailings leachate, Nelson, Reisenauer and Gee (1980) were able to use a value much like a retardation factor to describe the movement of an acid pH front. Both laboratory and field observations (Gee et al. 1980, Markos and Bush 1980, Relyea et al. 1982) have shown that contaminants in acidic leachate (pH less than 4) behind this front move at close to the velocity of ground water, but those contaminants in the neutral pH region (just ahead of the acid pH front) move very slowly or not at all compared to ground water.

2.4 SITE EVALUATION

The decision tree approach for determining whether or not a liner is needed, requires considerable characterization work. They include 1) enough site characterization to model water movement, 2) tailings characterization for determining the contaminant source term, and 3) characterization of the interactions of the leachate with soil and native ground water so that contaminant transport can be predicted. To obtain and utilize this information, a decision tree has been developed and is discussed at length in the following section.

3.0 DECISION TREE FOR LINER EVALUATION

The decision tree (Figure 2) is a logical sequence of questions to be answered and instructions to evaluate whether a particular site complies with the regulatory criteria for disposal of uranium mill tailings. It is intended to aid the firm(s) responsible for site design and remedial action of tailings piles exhibiting potential groundwater problems.

3.1 SITE CHARACTERIZATION

If the preliminary site assessment identifies a possible groundwater problem, decision tree methodology may be employed. The first step in the decision tree procedure is site characterization. Because of the initial expense for using liners or having to take further remedial action at a later date, comprehensive site characterization should be completed prior to construction activities to permit utilization of the decision tree in determining whether site specific objectives will be met. This saves both time and money if the design is insufficient to satisfy the criteria or if a more economical design that satisfies the criteria can be found.

It is possible (but unlikely) that sufficient data already exists to describe the site for the purpose of the decision tree. This is the first decision point in the tree: has the site been adequately characterized? If not, a site characterization plan should be devised to insure that all the needed data will be obtained.

The decision on liner emplacement will ultimately be based on predictions made with state-of-the-art computer models. These predictions will be only as good as the data obtained during site characterization. For this reason, the site characterization plan should be reviewed by an experienced hydrologist and geohydrochemist before execution of the site characterization plan.

FIGURE 2. Decision Tree Evaluation of Inactive Uranium Processing Sites for Liner Requirements. (The use of hydrologic and solute transport computer codes is implicit in the Decision Tree.)

Site characterization data may be divided into three categories:

1. Site Hydrology
2. Tailings Characterization
3. Contaminant Attenuation

Each of these will be discussed below.

3.1.1 Site Hydrology

Characterization of the hydrology can be considered sufficient if enough data exists to set up and run a hydrologic simulation that predicts the long term behavior of water at the site. More than one type of model may be required for the simulation, depending on the particular site.

Three sources of water exist that can remove contaminants: 1) infiltrating surface water, 2) subsurface or ground water, and 3) water that exists initially in the tailings. All three sources should be characterized to predict the behavior of a disposal site.

The water balance for surface water at a hypothetical site is shown in Figure 3. Precipitation and irrigation provide water to the surface, while runoff and evapotranspiration remove surface water. The water remaining is infiltration, which penetrates the cover layer of soil or fill. A layer of relatively impermeable material such as a cover to inhibit radon emission may divert some of the infiltrating water so that the net flux to the tailings is considerably lower than the infiltrating moisture. However, if radon is not considered a health hazard at a particular site, such a cover may not be in use.

Generally, an unsaturated flow model is used to predict the flux of water entering the tailings pile. Such a model (UNSAT1D) was applied to a similar situation by Bond, Freshley, and Gee (1982). Climatic data^(a) provide the time varying input (precipitation) to the UNSAT1D model as well as information

(a) These data may be obtained from:
The U.S. Department of Commerce
National Oceanic and Atmospheric Administration
Environmental Data and Information Service
National Climatic Center
Federal Building
Asheville, North Carolina 28801

P = PRECIPITATION, R = RUNOFF, ET = EVAPOTRANSPIRATION
 I = INFILTRATION, DI = DIVERTED INFILTRATION, F = FLUX INTO
 TAILINGS.

FIGURE 3. Water Balance for a Hypothetical Uranium Mill Tailings Disposal Site
 (Below Grade Example)

needed to calculate evapotranspiration. Such data include, for example, daily temperature fluctuations, wind speed, cloud cover and thunderstorm events (amount of rain, time of day, and duration). Calculation of evaporation also requires data describing initial moisture content, porosity, bulk density, and depths of surface and underlying soil or radon barrier layers. Hydraulic conductivities of soils and barrier layers are needed for unsaturated conditions (either unsaturated conductivities as a function of moisture potential or a saturated hydraulic conductivity along with a moisture retention curve). Finally, information on the type of vegetation and its water extraction behavior are needed.

For the decision tree, it must be assumed that the cover design has been completed and that the previously mentioned climate, vegetation and soils data are available or can be obtained. A complete description of unsaturated flow models for calculating the moisture flux into and through the tailings (Bond et al. 1982) is beyond the scope of this document. An experienced hydrologist or soil physicist should be consulted to ensure that 1) the model used to calculate infiltration and flux to the tailings is satisfactory, and 2) the data required for model calculations are adequate.

Once the water flux above the tailings pile has been calculated, it may be used as an upper boundary condition for an unsaturated flow (or combination unsaturated and saturated flow) model of the tailings and subsurface hydrology. The advantage of this approach is the difference in computational time steps required for a model which calculates evapotranspiration on a daily basis and a model that can use the infiltration/flux calculations on a monthly or yearly basis. The latter model can be run with longer time steps (thus shorter run time). The flux calculations from UNSAT1D for an average or maximum rainfall year are then input to a model which describes water transport through the tailings and subsurface layers. One model with this capability is TRUST (Reisenauer et al. 1982).

TRUST may be used to model subsurface water migration in one, two or three dimensions. It can also be used to model combined unsaturated and saturated flow. Initial and boundary conditions needed for this and other suitable models are similar.

Data required for unsaturated subsurface hydrologic models are:

- Tailings, soil and liner hydraulic properties including bulk density, porosity, saturated hydraulic conductivity and a moisture retention curve or hydraulic conductivity as a function of moisture potential. These may change as leachate makes contact with the soil and/or liner; therefore, multiple sets of measurements under different conditions may be needed.
- Initial moisture content, the piezometric surface, distribution and the location of each layer (e.g., depth of top and bottom of the layer) and boundary conditions at the maximum depth (e.g., depth to water table or free drainage at maximum depth).

If saturated flow is also to be considered (depending on the specific site), the depths, thicknesses and hydraulic conductivities of all layers must be known.

The data described in this section are all that are needed for the first test to determine whether or not a liner is required to satisfy design objectives: does water from an unlined pit stay within the site boundary? However, to make certain that all questions regarding contaminant release from a tailings disposal site are answered, geochemical characterization of tailings and the chemical interactions between tailings leachate and soil/liner materials and native ground water should be conducted simultaneously with site hydrologic characterization. This is logical from an economic standpoint because the subsurface and liner material samples needed for hydrologic characterization are also required for characterizing leachate interactions with these materials.

One important interaction that is needed for the hydrologic model is the behavior of liner and subsoil hydraulic conductivities as they are contacted with leachate. Gee et al. (1980) and Relyea et al. (1982) have measured hydraulic conductivities with tailings leachate in laboratory soil columns. Many of the saturated hydraulic conductivity values are observed to decrease as leachate is passed through the column. Also, characterization of the inactive

tailings may indicate that not all tailings require excavation and burial. Therefore, we will next discuss the parameters that need to be investigated to characterize tailings for the decision tree.

3.1.2 Tailings Characterization

Chemical compositions of uranium mill tailings may vary widely depending on the composition of the ore, the process used to extract uranium from the ore and what has happened to the tailings since their disposal. These three factors largely determine the quantities and rates of contaminants released from tailings upon contact with water. Knowing the leach rate of contaminants from a tailings pile, however, helps predict the behavior of a tailings disposal site.

Various investigations have been conducted that attempt to determine the quantities of contaminants that will be released from tailings (Markos and Bush 1981a and b; Markos, Bush and Freeman 1981; and Buelt et al. 1981). These studies used batch or jar tests to obtain tailings leachate for chemical analysis. The batch method involves contacting a tailings sample with water in a container and shaking for a period of time. Afterwards, the solid and liquid are separated and the liquid analyzed. Solid to solution ratios ranged from 1:5 to 1:100 (g tailings/cm³ water).

We have found that the batch method is acceptable qualitatively for screening a large number of tailings samples for relative contaminant concentrations, but it will not accurately determine how much contamination will be released from tailings and yields little (if any) information about contaminant release rates. Batch leaching tests give results that are crude averages (because of dilution) of contaminant concentrations over a larger number of pore volumes. For example, a 1:5 solid to solution ratio in a batch test is the same as contact between 17 pore volumes of water with a tailings pile having a bulk density of 1.5 and a porosity of 30%. Dilution also raises the pH in a batch test (compared to column leach tests), which limits the solubility of many elements. Thus, batch leach tests generally underestimate initial contaminant concentrations that may be leached from tailings upon contact with water. We have taken several tailings samples from three sites: Salt Lake City, Utah; Durango, Colorado; and Shiprock, New Mexico. These samples were

initially leached using a 100:1 batch method (Buel et al. 1981) and later using a 5:1 batch leach method. The 5:1 batch leaching results were used to select three samples for further tests with the column leaching method.

Table 1 shows a comparison of the batch leaching method with results from column leaching tests on the same tailings samples. The column leach test gives a much better estimate of leachate composition that would be released over time from a tailings pile.

Tailings leaching tests results from small columns (about 200 cm³ per column volume) have shown (Table 2) that most nonradioactive contaminants are released in the first one or two pore volumes of leachate. Contaminant concentrations of Al, As, Cd, Cr, Cu, Fe, Mg, Mn, Mo, Ni, P, SO₄, and Zn are highest in the first increment of leachate from the three tailings samples tested and decrease rapidly (generally by an order of magnitude or more) at the end of one pore volume. The behavior of radionuclides (²³⁸U, ²³⁵U, ²³⁰Th and ²²⁶Ra) is similar to that of nonradioactive elements as concentrations decrease by at least an order of magnitude after one pore volume.

Concentrations of As, Ba, Cr and Pb in leachate from acidic Salt Lake and Shiprock tailings were near to or below the EPA's proposed maximum concentration limit (MCL) levels (40 CFR Part 192) after less than two pore volumes. Similarly, leachate levels of Ba, Cd, Cr and Pb from alkaline Durango tailings decreased to below MCL after one or two pore volumes. Radioactive element concentrations after one pore volume were still observed to be at least three orders of magnitude above MCL levels.

Detection limits for the radioactive elements in leachate from small columns were found to be quite large, 10² to 10³ pCi/l. Therefore, large columns (column volume about 2000 cm³ and pore volume 1000 cm³) were subsequently used to obtain larger leachate samples for counting radioactive elements. As expected, detection limits were reduced by an order of magnitude.

The use of large columns allows analysis of both radioactive and nonradioactive contaminants in the same sample with the precision needed to critically assess the first tailings leachate pore volume. Our results indicate that effluent (leachate) from the tailings leach column should be collected in three

TABLE 1. Column-Batch Leach Test Comparison: Chemical Composition of Tailings Leachate (elemental concentrations in mg/l)

	Salt Lake		Shiprock		Durango	
	1:5 Batch	Column (a)	1:5 Batch	Column	1:5 Batch	Column
Al	4.0	1200.0	66.0	2700.0	0.6	11
Ca	600	550.0	560.0	530.0	1.1	7.2
Cd	0.05	2.6	0.1	5.3	<0.01	0.15
Co	1.0	37.0	0.3	11.0	ND ^(b)	0.03
Cu	1.9	48.0	3.5	130.0	ND	ND
Fe	6.7	1700.0	2.4	160.0	ND	0.17
K	4.9	ND	4.2	66.0	7.8 ^(c)	43.0
Mg	14.3	280.0	54.0	2100.0	7.2	17.0
Mn	2.2	55.0	1.5	65.0	ND	0.02
Na	5.5	110.0	31.5	1500.0	120.0	1900.0
Ni	0.6	21.0	0.3	11.0	ND	ND
Si	33.1	138.0	25.9	79.0	6.1	8.5
Sr	8.4	14.0	2.6	9.2	0.2	0.08
Zn	2.2	98.0	2.9	120.0	ND	0.12
pH	3.0	2.0	3.1	2.3	9.3 ^(c)	9.5
E.C.	2900	12,000	3250	23,000	380 ^(c)	6300
(micro-mho/cm)						
<u>Column Data</u>						
Bulk Density (g/cm ³)	1.21	1.35		1.30		
Porosity (cm ³ /cm ³)	0.56	0.48		0.49		
Pore Water Velocity (cm/hr)		0.41		0.56		0.48
Column Length (cm)	25.4	25.4		25.4		

(a) The volume of effluent collected corresponds to the first 0.20 to 0.27 pore volumes for the leach tests.

(b) ND = Not Detected in the sample.

(c) The pH electrode broke in this sample, so values for pH, K and E.C. were taken from an earlier analysis. KCl from the electrode are not expected to affect analysis of other elements.

TABLE 2. Tailings Leachate Compositions in the Initial 0.25 Pore Volume of Effluent from Column Leaching Experiments

		Concentrations in Leachate From Tailings Sample (mg/l)		
		Shiprock	Salt Lake	Durango
Al	2,700	1,200	11	
Ca	530	550	7.2	
Cu	130	48	ND ^(a)	
Co	11	37	0.03	
Fe	160	1,700	0.17	
K	66	ND	43	
Mg	2,100	280	17	
Mn	65	55	0.02	
Na	1,500	110	1,900	
Ni	11	21	ND	
Sr	9.2	14	0.08	
Zn	120	98	0.12	
B	ND	ND	4.9	
Cl	120	48	360	
CO ₃	ND	ND	1,200	
F	14	470	14	
Mo	ND	9.7	2.5	
NO ₃ +NO ₂	750	ND	7	
P	ND	64	25	
SO ₄	34,000	13,000	900	
Si	79	140	8.5	
As	11	12	16	
Ba	0.1	ND	0.03	
Cd	5.3	2.6	0.15	
Cr	1.9	2.9	1.6	
Pb	ND	ND	ND	
²³⁵ U (pCi/l) ^(b)		320	3,900	86
²³⁸ U	6,800	83,000	1,850	
²³⁰ Th	38,500	280,000	ND	
²²⁶ Ra	2,600	31,000	1,200	
²¹⁰ Pb	670	ND	ND	
pH	2.3	2.0	9.5	
Electrical Conductivity (mmho/cm)		23	12	6.3

(a) ND = Not detected.

(b) Radionuclides were analyzed with samples from large tailings columns; pore volumes 1,000 cm³.

or four equal fractions for analysis of the first pore volume. The second and third pore volumes should be collected in at least two fractions and thereafter sampled at least once per pore volume of leachate. Figure 4 shows the pH and E.C. (electrical conductivity) measurements for leachate from a sample of tailings from the Salt Lake City site. For this sample (and those from Durango and Shiprock), only gradual changes were observed in both pH and E.C. after sampling four to five pore volumes of leachate. The gradual change indicates that leachate composition changes very little after ten pore volumes. Therefore, very little information is gained by analyzing tailings leachate after that point.

Buelt et al. (1981) and Markos and Bush (1981a, 1981b) have used the batch leach test to screen many tailings samples from each of several sites. Their results have shown a great deal of variability in the tailings leachate from each site. Because of the ease and speed with which a batch test can be conducted, it is valuable for characterizing the variability of a tailings pile. The batch test results can be used to select a representative number of tailings samples for further testing with the column leaching method. The results from column leaching tests should then be used to estimate the leachate composition expected to be released from the tailings pile. At this point hydrodynamic dispersion, dilution, chemical precipitation and adsorption represent the second line of defense against significant contamination of surface or ground water. Estimation of leachate composition from a large number of samples is a statistical problem in spatial variability that is beyond the scope of this project; therefore, we will use "worst case" leachate for the decision tree demonstration (e.g., leachate having properties such as extremes in pH and high contaminant levels).

The contaminant source term generated by characterization of uranium mill tailings samples with the column leaching test is used as a boundary condition for contaminant concentrations at the tailings-soil or tailings-liner interface. Without data to describe the interactions of leachate with soil and liner material we must assume that contaminants migrate unattenuated with the same velocity as water through subsurface layers. This assumption is clearly in error and can lead to expensive and potentially unnecessary engineered

FIGURE 4. pH and Electrical Conductivity Measurements for Effluent from Column Leaching Experiments with Tailings from the Salt Lake Site

barriers being constructed. To be able to make realistic predictions of attenuated contaminant migration, the interactions of tailings leachate with soil and liner material must also be quantified.

3.1.3 Contaminant Attenuation

In order to proceed with the decision tree to the point that predictions of retarded (or attenuated) contaminant concentrations at a site boundary can be compared with regulatory criteria, more data must be obtained for the hydrologic model. These data must somehow link the migration of contaminant to the predicted water movement. This common link between solute (contaminant) and water transport is the retardation factor, which is discussed briefly in Section 3.0 and in more detail by Relyea, Serne and Rai (1980) and Relyea (1982).

Generally, the retardation factor is valid under very restricted, ideal conditions where only adsorption is considered. A discussion of recent investigations of solute retardation under less than ideal conditions in saturated porous media is given by Nkedi-Kizza et al. (1982), but the only mechanism for solute retardation in this transport model is ideal adsorption (e.g., linear adsorption isotherm). A solute transport model for retardation by non-linear adsorption in both saturated and unsaturated porous media is given by Persaud and Wierenga (1982). Neither of these two transport models (or any other model) is presently capable of adequately predicting solute/contaminant transport under the conditions found at a uranium mill tailings disposal site. The reason for this is that state-of-the-art solute transport models are not yet capable of describing contaminant attenuation caused by precipitation, contaminant scavenging by coprecipitation and nonideal adsorption. This limitation results in overly conservative predictions since most contaminant attenuation from acidic tailings piles is apparently a result of pH induced precipitation reactions (Gee et al. 1980, Markos and Bush 1981b).

Fortunately, contaminant precipitation from tailings leachate is very dependent on pH, which can be predicted as the leachate moves through subsurface soil and liner material (Nelson 1980). To then predict contaminant migration, two retardation factors are required for each contaminant: one retardation factor to describe contaminant migration in unneutralized (low pH) leachate and another that describes contaminant migration through soil or liner

in neutralized (higher pH) leachate. Two retardation factors are needed for each contaminant in each layer of soil or liner material that contact the leachate. A retardation factor for the pH is also needed. Methods for obtaining the required data are discussed below.

pH Retardation Factor

The retardation factor R_f for an ideally adsorbed contaminant is given by

$$R_f = 1 + \frac{\rho_b}{\theta} R_d \quad (1)$$

where

ρ_b = bulk density of the porous media

θ = the volumetric water content of the porous media; under saturated conditions θ is equal to the porosity ϵ

R_d = the adsorption coefficient (or distribution ratio) for the contaminant.

The distribution ratio, R_d , is defined by

$$R_d = \frac{\text{Amount of contaminant adsorbed on the solid (meq/g)}}{\text{Amount of contaminant in solution (meq/cm}^3\text{)}}$$

The R_d (or K_d , as it is sometimes referred to) can be measured using either batch or column techniques (Relyea et al. 1980, Relyea 1982). We shall use the approach of Nelson et al. (1980) to define an R_d value for the special case of pH.

The titration curve, shown in Figure 5, gives the change in the pH of a Clive, Utah soil as leachate from Shiprock tailings is added. Our studies and those of Gee et al. (1980) have shown that many of the contaminant concentrations are greatly reduced at pH 7.7. Therefore, we shall find the R_d for the pH at pH = 7.7.

FIGURE 5. Titration Curve for Soil A from Clive, Utah with Shiprock Tailings Leachate

Acidity (high H^+ concentrations) may be thought of as a contaminant because highly acidic conditions increase the solubilities, and therefore concentrations, of many other contaminants. The total acidity for leachate is given by the sum of $H^+ + Al^{+3} + Fe^{+3}$ concentrations in meq/l. The total acidity includes Al^{+3} and Fe^{+3} in addition to H^+ because at higher pH values Al^{+3} and Fe^{+3} hydrous oxides are precipitated and release hydrogen ions equivalent to the original Al and Fe concentrations. From Table 2 the total acidity is found to be 300 meq/l. At pH 7.7 this soil neutralizes or adsorbs 230 meq total acidity per 100 g (see Figure 5). The resulting Rd for Shiprock leachate at pH = 7.7 in this soil is then

$$Rd = \left(\frac{230 \text{ meq/100 g}}{300 \text{ meq/1000 cm}^3} \right) = 7.7 \text{ cm}^3/\text{g}$$

The Rd obtained implies that 1 g of this soil will neutralize 7.7 cm^3 of Shiprock leachate to a pH of 7.7.

If we had chosen pH = 4.5 for the above example we would have found that 100 g of soil neutralizes 470 meq of acidity. The Rd value for pH = 4.5 is then

$$R_d = \left(\frac{470 \text{ meq}/100 \text{ g}}{300 \text{ meq}/1000 \text{ cm}^3} \right) = 15.7 \text{ cm}^3/\text{g}$$

One gram of soil will neutralize 15.7 cm of shiprock leachate to pH 4.5.

The retardation factor for this soil can be found from Equation (1). If the soil bulk density is $1.5 \text{ mg}/\text{cm}^3$ and porosity 0.4, then the pH 7.7 retardation factor is

$$R_f = 1 + \frac{\rho_b}{\theta} R_d$$

or

$$R_f = 1 + \frac{1.5}{0.4} (7.7) = 30$$

This means that the Shiprock leachate pH front (at pH 7.7) moves 30 times slower than the water under saturated conditions.

The pH retardation factor at pH = 4.5 is

$$R_f = 1 + \frac{1.5}{0.4} (14.7) = 56$$

The Shiprock leachate pH front at pH 4.5 moves 56 times slower than water.

Contaminant Retardation Factors

To predict contaminant transport, we must find two Rd values for each contaminant. For this example one Rd value is needed at pH 7.7 and another is needed for the contaminant in unneutralized leachate. Gee et al. (1980) used a batch method to calculate Rd values. A known volume of leachate (V) was added to a known amount of soil (W). Contaminant concentrations were measured before contact (C_0) and afterward (C_f). The amount of contaminant adsorbed on or precipitated in the soil is $(C_0 - C_f)V$. Therefore, the contaminant concentration

per gram of soil is $(C_0 - C_f)V/W$. The R_d is defined by the ratio of the contaminant concentration per gram of soil to the final contaminant concentration in the leachate:

$$R_d = \frac{(C_0 V - C_f)/W}{C_f}$$

or more simply,

$$R_d = \frac{V}{W} \left(\frac{C_0}{C_f} - 1 \right) \quad (2)$$

Two R_d values are required for each contaminant (one for unneutralized and one for neutralized tailings leachate). An R_d value is also needed to describe movement of the pH front. If alkaline or neutral tailings are leached, precipitation may not be a major retardation mechanism. In these cases, only one R_d value may be required (for each contaminant). Once the site has been characterized, the first box in the decision tree is complete and we may proceed to the second decision point.

3.2 WATER MOVEMENT WITHOUT A LINER

This step requires an experienced ground-water hydrologist who is able to interpret and utilize the site characterization data to set up a water transport model that describes the disposal site. This step is far from trivial, but details are beyond the scope of this document. Readers are referred to Simmons and Gee (1981), Reisenauer et al. (1982), Nelson et al. (1980) and Bond et al. (1982) for more detailed information on setting up and running computer simulations of water transport.

Once the computer model or models have been set up and run, the question is: Does water from the tailings stay within the site boundary during the control period as defined by state and federal regulations? If water does stay within the site boundary for an unlined disposal area, then contaminants will also remain within the boundary. However, the computer model is already set up at this point and contaminant transport can be predicted for one fourth to one third the cost of setting up and running the water transport simulation. Although a prediction of contaminant transport may not be required, it may be

very useful as additional evidence that a site without a liner more than satisfies the regulatory criteria. If water from the unlined tailings pit is found to move beyond the site boundary, then contaminant transport must be predicted.

3.3 CONTAMINANT TRANSPORT WITHOUT A LINER

At this point, water is predicted to move from the tailings to beyond the site boundary at some time less than a specified control period. The next step is to predict contaminant migration using the tailings leachate data and contaminant retardation data gathered during site characterization. The question is: do contaminant concentrations from the tailings stay below the limit set by the disposal site design objectives at or beyond the site boundary for an unlined disposal area? If the answer is yes, then no liner is required for tailings disposal at that site. If the answer is no, then contaminant levels above the limits are predicted to cross the boundary for an unlined tailings pile; thus, a new disposal design must be formulated with the possible inclusion of a liner, or new design objectives must be established in conformance with regulatory agencies.

3.4 WATER MOVEMENT WITH A LINER

The water transport model for the site may be altered to include a liner at this point. Liner thickness and hydraulic conductivity are added to the water transport model. A new computer simulation of water movement with the liner must be run and compared with the design objectives. The question is now: does water from a lined tailings disposal area stay within the site boundary in the control period specified by the design objectives agreed to by state and federal agencies? If the answer is yes, then a lined tailings disposal area satisfies the objectives and may be used. However, the cost of a liner system must be affordable in relation to the beneficial use of the groundwater described by the design objectives. Again, the prediction of contaminant migration may be useful as additional evidence that the site design is satisfactory. If the answer is no, then water from tailings is predicted to cross the boundary and the movement of contaminants must be considered.

Various liner materials may be evaluated at this point. Barnes et al. (1981) have published data on the performance of catalytic airblown asphalt membranes. Recent data indicate permeabilities in the range of 10^{-11} to 10^{-12} m/s. Sodium bentonite amended soils may also be used for low permeability liner material.

3.5 CONTAMINANT TRANSPORT WITH A LINER

Contaminant attenuation data (contaminant R_d values, R_d value for the selected pH front) for the liner and subsurface soils must now be added to the water movement model for a lined disposal site. Another computer simulation is required to answer the question: do contaminant concentrations transported from a lined tailings disposal area stay within the site boundary during the control period? If contaminant concentrations at the site boundary meet the objectives during the control period, then a lined tailings disposal site can be used. Again, the cost of the liner system must be taken into consideration. If the objectives are not met, predicted contaminant releases are excessive and the site should be redesigned (by using a thicker liner, different liner material or a new approach) or an alternative site considered. Both of these options require that the decision tree process be repeated from the start. If the same site is to be used with a new design, however, much of the characterization is already completed.

4.0 DEMONSTRATION OF DECISION TREE: EXAMPLE CASE

For the demonstration of the decision tree, data from several sites will be utilized. The demonstration is not an evaluation of any particular site or any specific tailings disposal method. It provides two example cases meant to aid the reader in determining the requirements to conduct a site evaluation for liner requirements.

4.1 SITE CHARACTERIZATION

4.1.1 The Existing Site

Preliminary data for a draft Environmental Impact Statement (EIS) on the Clive, Utah site was obtained by personal communication from Martin Tierney of Sandia Laboratories in Albuquerque, New Mexico. The interpretation of this data has been simplified to eliminate excessive detail which might otherwise detract from the main points of the demonstration.

The Clive, Utah site is a 259 ha (1.0 mi²) area in Tooele County about 105 km (65 mi) west of Salt Lake City. The site slopes gently from the NE to the SW with a maximum drop of 3.35 m (11 ft) within the area. Vegetation consists of sparse brush and weeds.

Surface and subsurface soil conditions were evaluated through the drilling of 13 exploratory bore holes. Detailed logs were kept and several samples were obtained for each borehole. Laboratory measurements on samples were made for particle size, permeabilities and other physical properties. Ground-water levels were measured periodically for seven months for five boreholes and over a two month period for the remaining eight boreholes.

A simplified concept of the existing stratigraphy (W to E) in the middle of the Clive, Utah site is shown in Figure 6. The subsurface layers generally rise from W to E while the ground-water level drops slightly. The surface rise from W to E is about 1.22 vertical m (4 ft) in 610 horizontal m (2000 ft), and 0.15 m (0.5 ft) or less in 610 m (2000 ft) from S to N. The ground water hydraulic gradient is approximately 4×10^{-4} m/m from the SW to the NE. If we

SOIL A: SILTS AND CLAYS, LIQUID LIMIT LESS THAN 50
 SOIL B: SANDY SOILS, MORE THAN 50% LESS THAN 5mm DIAMETER

0 50 100 m
 HORIZONTAL
 SCALE

FIGURE 6. Simplified Stratigraphy for the Clive, Utah Site

assume that saturated hydraulic conductivities below the water table are on the order of 10^{-5} m/s, the ground-water velocity would be 10^{-8} m/s (1.0 ft/yr) from SW to NE with a porosity of 40%.

Two types of soils will be assumed for the demonstration. Soil A in Figure 5 ranges from a clayey silt to a silty clay with a saturated hydraulic conductivity of 10^{-9} m/s. Unsaturated hydraulic conductivities for this soil are assumed to be similar to those of a shale packed to a bulk density of 1.5 mg/m^3 reported by Bond et al. (1982). Soil B is a sandy soil with saturated hydraulic conductivity ranging from 10^{-7} m/s to more than 10^{-5} m/s. We will assume that this soil has the same hydraulic properties as the soil properties used by Bond et al. (1982) [i.e., saturated conductivity equal to 10^{-6} m/s (100 ft/yr)]. Soil and tailings hydraulic properties needed for the computer model are given in Appendix B.

Climatic data were taken from Grand Junction, Colorado, which has a climate and vegetative cover somewhat similar to that at Clive, Utah. These data were monthly summaries for 1979 obtained from the National Oceanic and Atmospheric Administration (NOAA). Total precipitation for Grand Junction in 1979 was 0.307 m (12 in.). A more detailed summary of these data is reported by Bond et al. (1982).

4.1.2 Hypothetical Design for Disposal of Tailings

To obtain any further data for the hydrologic simulation, a design for the disposal site is needed. The depth of cover layers, liner and tailings and their hydraulic properties are needed. Unsaturated conductivity and other hydraulic properties for the tailings are taken from Nelson et al. (1980). Depths of the cover layer are shown in Figure 7. Approximately $2 \times 10^6 \text{ m}^3$ of tailings from inactive mills are presently stored at each of three sites: Salt Lake, Shiprock and Durango. The simple disposal system shown in Figure 6 is large enough to accommodate all the tailings from any of those sites. The pit dimensions are: depth = 6.1 m (20 ft), width = length = 580 m (1900 ft). The surface of the pit would cover ~32 ha (80 acres). About 1.8 m (6 ft) of the sandy soil B would be found below the pit and the depth to water table below the pit ranges from 1.52 to 3.35 m (5 to 11 ft).

FIGURE 7. Simplified Stratigraphy for a Hypothetical Unlined Disposal Site at Clive, Utah

The cover would consist of two 1.52 m (5 ft) layers of soil from the site: top soil A as an overburden layer adjacent to the tailings and sandy soil B for the final cover. Hydraulic properties for these layers are assumed to be unchanged by excavation, repacking, and revegetation.

The case for a lined pit is shown in Figure 8. Here the pit is excavated to 7.0 m (23 ft) below grade and backfilled with soil A to a depth of 6.1 m (20 ft) (liner thickness of 0.9 m or 3 ft). Again, it is assumed for simplicity that repacking soil A for the liner will have no effect on its hydraulic properties.

4.1.3 Contaminant Source Term

The data described thus far completes site characterization for the purpose of hydrologic modeling for both conditions (with and without a liner), but data are still needed for making predictions about contaminant transport. The first data required for prediction of contaminant behavior is the contaminant source term (or tailings leach test results).

Figures 9 and 10 show pH and electrical conductivity measurements for Shiprock and Durango tailings samples. These tailings represent the extremes for chemical conditions found in our laboratory studies (Relyea et al. 1982). Durango tailings leachate is an alkaline leached sample; therefore, its leachate has a high pH but relatively low total dissolved solids. The pH is observed to drop slowly from near pH 10 down to about pH 8, thus buffering of the leachate pH by soil and liner material is not expected to be a major factor in contaminant attenuation (because the soil pH and leachate pH values are not much different). The electrical conductivity and contaminant concentrations in Durango tailings leachate fall rapidly as water is leached through the tailings. Radium and uranium concentrations, however, are still three orders of magnitude above MCL levels after one pore volume and As is also a factor of ten above its MCL.

The Shiprock tailings leachate represents an acidic leachate with high total dissolved solids. Again, contaminant concentrations and electrical conductivity fall rapidly as the tailings are leached. Neutralization and associated precipitation occur when leachate contacts soils or liner material and are expected to greatly reduce contaminant concentrations. For the

FIGURE 8. Simplified Stratigraphy for a Hypothetical Lined Disposal Site at Clive, Utah

FIGURE 9. pH and Electrical Conductivity Measurements for Effluent from Column Leaching Experiments with Tailings from the Durango Site

demonstration we will assume that leachate concentrations within the tailings pile are constant over time and space and are represented by the analyses for Shiprock and Durango leachate given in Table 2. These are "worst case" conditions and would be modified to show that leachate contaminant concentrations

FIGURE 10. pH and Electrical Conductivity Measurements for Effluent from Column Leaching Experiments with Tailings from the Shiprock Site

decrease with time as the source of contaminants is gradually used up. In an actual exercise with the decision tree, these decreases of leachate contaminant concentrations would be modeled to reflect observed changes in the leachate composition.

4.1.4 Contaminant Attenuation

The distribution ratio, R_d , for pH 7.7 with soil A was calculated in Section 3.0 for Shiprock tailings leachate. The R_d value for pH 7.7 with soil B and Shiprock leachate was estimated from leachate pH breakthrough curves for Morton Ranch leachate and overburden reported by Gee et al. (1980). No distribution ratio for pH was calculated for Durango leachate because it is alkaline and very little (if any) neutralization occurred in batch or column tests. Batch R_d values on soil A were measured for As, Cd, Cr and Pb with Shiprock leachate and for As, Cd and Cr with Durango leachate. These values were measured by shaking 250 cm³ of leachate with 50 g of soil A for two weeks and analyzing the resulting solutions. Initial and final contaminant concentrations were used to calculate an R_d for each element using Equation (2). The Shiprock R_d values for Ra, U and Th on soils A and B at pH 7.7 were also taken from Gee et al. (1980) for those elements in Morton Ranch tailings solution on clay liner material. R_d values for contaminants at pH 7.7 on soil B were assumed to be a factor of ten less than the values for soil A. The R_d values for As, Cr, and Cd at pH < 7.7 were assumed to be zero similar to the results of Gee et al. (1980) for unneutralized leachate.

An additional contaminant attenuation mechanism that is not utilized for this demonstration is the decrease in soil and clay liner permeability often observed when they contact tailings leachate (Gee et al. 1980, Relyea et al. 1982). This phenomenon may be modeled by decreasing the liner or soil permeability in steps as known pore volumes of tailings leachate are released. This may result in the release of less leachate from the tailings over time. However, the leachate may then fill up the tailings pit and migrate to the surface or spill over the sides as in an overfull bathtub. For the demonstration we will assume that permeability values are constant; however, in an actual site evaluation the permeability decrease must be included in the transport model.

Site characterization is complete when all data needed to predict behavior of the site are obtained. The first predictive step is to set up the hydrologic model (or models) and simulate water movement over time.

4.2 RESULTS WITHOUT A LINER

The design selected for tailings disposal for this demonstration was described in the previous section. The burial pit is shallow (6.10 m) because of the relatively high water table. To accommodate two million cubic meters, the pit must cover 32 ha (80 acres). The ratio of the horizontal to vertical dimension of the pit is about 100:1; therefore, we can simulate water movement through the tailings pit with a one dimensional model.

An unsaturated flow model UNSAT1D (Bond et al. 1982) was used to calculate evapotranspiration and infiltration with the climate data from Grand Junction, Colorado and the pit cover design shown in Figures 6 and 7. Rainfall was predicted to readily infiltrate the sandy soil postulated for the cover. Of the 0.30 m annual precipitation, 0.256 m is predicted to return to the atmosphere either by evaporation or through the transpiration of cover vegetation. The remaining 0.044 m passes through both the sandy layer (soil B) and the clay cover (soil A) to contact the tailings. Most of the 0.044 m of water passing through the cover layers was a result of a few large rainstorms. Water from these few events infiltrated below the very sparse vegetative cover root zone and thereby escaped surface effects such as transpiration and evaporation. This demonstrates why monthly or yearly average precipitation and evapotranspiration values can be quite misleading. Because of the scarcity of plants at the site, we assumed that no plant cover exists and water is returned to the atmosphere only by evaporation.

4.2.1 Water Movement

Infiltration through the cover predicted by UNSAT1D (0.044 m/yr) was used as the upper boundary condition for TRUST at the cover tailings interface. TRUST was used with a time step of 1 day to simulate unsaturated flow through the tailings and underlying soils to the water table. The 1 day time step greatly reduces computer time compared to UNSAT1D run times with hourly time steps.

Initial moisture contents for the various layers (covers, tailings and subsoil) were chosen based on past experience and the data reported by Gee et al. (1980) and Bond et al. (1982). TRUST was then used to simulate water movement until a steady state is reached (water moving downward into the tailings; 0.044 m/yr equals the amount of water moving out the bottom to the water table). The moisture contents of the clay cover layer (soil A) was found to be 98% saturated (i.e., 98% of the porosity is filled with water), while the subsoil (soil B) was 46% saturated at the subsoil-tailings interface and increased to 100% saturation at the water table. At steady state, the tailings were 51 to 52% saturated.

For the simulation of actual water movement through the tailings, it was assumed that the tailings would initially be somewhat drier than 51% because of draining and evaporation during the period in which the tailings are excavated, moved and placed into the pit. A value of 30% saturation was selected for the initial moisture content of the tailings.

The cover layer of soil A was assumed to be at 98% saturation initially because, as a barrier to radon gas emission, field tests have shown that radon emission is greatly reduced by cover layers with low air filled porosity (Nelson, Gee and Oster 1980). Therefore, it is likely that such a cover layer would be initially wet to reduce radon emission. Subsoil between the tailings and water table was assumed to have the same moisture content obtained under steady-state conditions.

With the initial conditions described above, the TRUST water movement simulation showed that water would move both upward from below the tailings and infiltrate downward into the unsaturated tailings pile. After 36 years the tailings begin to drain by unsaturated flow to the water table. After 51 years water that was initially in the bottom of the tailings pit would reach the water table. The 51 year travel time will be used for calculation of water transport time to the site boundary. For this demonstration we will assume that no dilution occurs and the water from the tailings pit moves with the ground water at 0.305 m/yr to the NE. After approximately 328 years, water from the edge of the pit will have traveled 100 m which is arbitrarily named the site boundary. Total travel time for water from the pit to the site boundary is 379 years (51 + 328). The answer to the first question in the decision

tree is NO; water from the pit does not stay within the boundary in the first 1000 years. This requires that we consider contaminant transport and proceed to the next step of the decision tree.

The 100 m boundary was established for purposes of demonstration only. It does not reflect recommended criteria for any site. Nor does the design objective of preventing contaminant transport to the site boundary reflect recommended design objectives. Actual design objectives for a site will be established on the basis of the potential beneficial use of the groundwater with concurrence of local, state, and federal agencies. If they include chemical concentration specifications, the effects of dispersion, dilution, and chemical attenuation would have to be included in the chosen models.

4.2.2 Contaminant Transport

We will consider two cases for contaminant transport: 1) contaminants from alkaline Durango tailings leachate, and 2) contaminants from acidic Shiprock tailings leachate. R_d values required to predict movement of selected contaminants in these leachates are given in Table 3. In both cases leachate would begin to leave the bottom of the pit as infiltrating water reaches the subsoil at about 36 years; however, the subsoil (soil B) would retard contaminant migration by adsorption and/or precipitation of those contaminants. When these contaminants reach the water table they would then move toward the site boundary. Travel times for water, the acid front and each contaminant to a 100 m boundary are given in Table 4.

4.2.2.1 Alkaline Tailings

For the contaminants As, Cd and Cr from Durango leachate, the R_d values for soil B in Table 3 are used to predict their movement from the pit bottom to the water table. Results of the computer simulation show the As and Cd fronts arrive at the water table in 94 years, while Cr lags behind at 134 years.

If we neglect the effects of dispersion and diffusion and assume there is no dilution of contaminants by ground water, the time required for these contaminants to reach the boundary (100 m) can be found from the retardation factor [Equation (1)] and the ground-water velocity. Under saturated conditions, the volumetric water content of the soil below the water table is assumed to be

TABLE 3. Rd Values and Initial Concentrations Used for the Decision Tree Demonstration

<u>Durango Tailings Leachate</u>					
Element	Rd (ml/g)		Initial Leachate		
	Soil A	Soil B	Concentration		
As	3.5	0.5	16.0 ppm		
Cd	1.0	0.5	0.15 ppm		
Cr	3.8	1.0	1.6 ppm		

<u>Shiprock Tailings Leachate</u>					
Element	Rd (ml/g) pH > 7.7		Rd (ml/g) pH < 7.7		Initial Leachate
	Soil A	Soil B	Soil A	Soil B	
pH	7.7	0.1	---	---	---
As	90	6	0	0	5.3 ppm
Cd	465	40	0	0	1.9 ppm
Cr	470	40	0	0	11.0 ppm
Pb	120	12	10	1.0	300 pCi/l
Ra	2,300	230	13	10	30,000 pCi/l
U	23,000	2,300	1.3	1.0	1,000 pCi/l
Th	80,000	8,000	1.2	1.1	100,000 pCi/l

0.44 and its bulk density is 1.5 g/cm^3 . With the Rd values for soil B from Table 3 we find a retardation factor R_f of 2.70 for As and Cd and $R_f = 4.41$ for Cr. Under the above assumptions, it would require 980 years after closing the pit for As and Cd to reach the boundary and almost 1600 years for Cr. The total time to reach the boundary is greater than 1000 years for Cr but not for As or Cd; thus the disposal design does not satisfy the objectives established for this case. However, As and Cd fall just short of satisfying the 1000 year criteria.

Dilution, diffusion and dispersion by the ground water are neglected in this analysis. A more realistic analysis of contaminant transport in the ground water would be possible for an actual site which has been adequately characterized.

4.2.2.2 Acidic Tailings

To predict contaminant transport from acidic tailings, we must first determine the R_d value of the pH front in the soils through which the leachate must flow. Results for batch interaction studies with soil A and Shiprock tailings leachate are shown in Appendix B. This soil buffered the pH at 7.7 for leachate initially at pH 2.1. Therefore, the pH value we have selected for this demonstration is 7.7. The R_d value for pH in soil B is an unknown because no soil samples were available. Permeability cell data from Gee et al. (1980) for Morton Ranch tailings solutions and overburden shows that an appropriate acid front R_d value for pH 7.7 is in the range 0.1 to 0.2. We have therefore assumed that soil B is similar (very little neutralizing capacity) and has an $R_d = 0.1$ for the acid front (pH = 7.7).

For the case of acid tailings in an unlined pit, leachate will move almost as fast as the infiltrating water. The elements As, Cd and Cr will move with the leachate because they are not slowed appreciably by adsorption or precipitation in the unneutralized leachate ($R_d = 0$ for As, Cd and Cr). They will, however, be slowed down ahead of the pH front (i.e., when the leachate is neutralized to pH > 7.7). Radioactive elements Pb, Th, U and Ra are slowed down relative to the movement of the pH front because their R_d values are assumed to range from 1.0 to 10 ml/g in unneutralized (acid) leachate.

Results from the computer simulation predict that As, Cd and Cr will arrive at the water table below an unlined pit after 61 years and at the 100 m boundary at 500 years (somewhat longer than the water arrival times in Section 4.2.1). The radioactive elements Pb, U and Th would arrive at the water table between 110 to 120 years. Total travel times for these elements to the 100 m site boundary range from 1100 years for Pb and U, 1500 years for Th, and up to more than 11,000 years for Ra.

The results of this analysis show that the radioactive contaminants Pb, Ra, U and Th do not exceed the site boundary after 1000 years. However, As, Cr and Cd would exceed the design objectives after 500 years. Assuming the design objectives are sustained after the cost analysis for the liner is performed, the present site design cannot be utilized for these acidic tailings without a

TABLE 4. Comparison of Arrival Times Predicted for Contaminants at a 100 m Site Boundary for Hypothetical Lined and Unlined Tailings Disposal Pits

	Arrival Time (yr)	
	<u>Unlined Pit</u>	<u>Lined Pit</u>
<u>Durango Tailings Leachate</u>		
As	980	1,049
Cd	980	991
Cr	1,600	1,647
<u>Shiprock Tailings Leachate</u>		
As	500	667
Cd	500	667
Cr	500	667
Pb	1,100	1,400
Ra	11,000	12,000
U	1,100	1,400
Th	1,500	1,800

liner or an alternate foundation soil (soil A). To evaluate a design requiring a liner, the next step of the decision tree requires that water movement with a liner be evaluated.

4.3 RESULTS WITH A LINER

To evaluate a lined tailings pit, the unsaturated water flow model (TRUST) used in Section 4.2 must be modified to include a liner that is 0.9 m thick made from soil A. The unsaturated hydraulic conductivity, initial moisture content, bulk density, porosity and liner thickness are the new input variables added to the previous model. Infiltration through the cover layer is assumed to be unchanged by the existence of a liner below the tailings. A computer simulation of water movement through the tailings, liner and subsoil to the water table was then performed.

4.3.1 Water Flow

As in Section 4.2.1, a water movement simulation was made to determine steady state conditions. The clay cover layer (a 1.5 m thick layer of soil A)

was found to maintain 98% saturation while the bottom clay liner (a 0.9 m thick layer of Soil A) was between 98.6 and 99.0% saturation. The tailings under steady state conditions were at 51% saturation. For the water transport simulation it was assumed that the clay cover layer and bottom clay liner were initially at their steady state moisture contents. The tailings were assumed to be drained to 30% saturation during the process of excavation, transport and unloading into the pit.

Results of the water movement simulation show that water from below the pit would initially move upward into the bottom of the tailings pile (as in the unlined case), while infiltration through the cover would move downward to the top of the tailings pile. After approximately 36 years the tailings would reach 51% saturation and begin to drain through the liner. Travel time to the water table for water that was initially in the bottom of the tailings pit was found to be 48 years. Movement of water over time from the bottom of the pit to the water table is shown in Figure 11. The travel times for water from the bottom of the tailings pile to the water table are not significantly different for the lined and unlined pits. Had we started the simulation with less than 98% saturation of the liner, less water would be in contact with the bottom tailings and more water would have to be drawn up from the water table. This would increase the time required before the tailings could drain, thereby increasing the travel times for water from the lined pit to the water table. However, the assumption that the liner is initially 98% saturated is probably not far from reality because during the period in which the pit is filled with tailings, rainfall would quickly move into the liner, and moisture from the water table would be drawn upward into the unsaturated clay liner.

With a travel time of 48 years from the bottom of the pit to the water table and a travel time of 328 years from the edge of the pit to the 100 m boundary we find that approximately 376 years are needed before water from the tailings reaches the site boundary. As far as the 1000 year objective is concerned, the soil liner we have used for the computer simulation makes very little difference in the movement of water to the site boundary.

If, on the other hand, a more impermeable soil liner (i.e., saturated K about 10^{-12} m/s or less) had been used in the simulation, water might possibly

FIGURE 11. Movement of Water from the Bottom of the Tailings (at 9.1 m) to the Water Table (11.5 m). (The dashed line indicates an unlined pit and the solid lines a lined pit.)

have accumulated in the tailings. The resulting saturated zone of tailings would form a perched water table above the liner, thereby increasing the moisture potential gradient across the liner. The depth of the steady-state saturated tailings zone would be determined by the infiltration rate and the saturated liner permeability. A one dimensional water flow model is not likely to be adequate for these cases because of the possibility of water movement through side walls of the pit. A case similar to this is analyzed by Nelson, Reisenauer and Gee (1980).

4.3.2 Contaminant Transport

4.3.2.1 Alkaline Tailings

To simulate contaminant transport for Durango tailings leachate and a soil liner, an R_d value is required for each contaminant. The R_d values for As, Cd and Cr on the proposed liner material (soil A) are shown in Table 3. The 0.91 m thick liner will not change travel times for the contaminants (compared to the unlined pit) once they reach the water table. The liner may, however, significantly increase the time required for the contaminants to reach the water table (see Table 4).

Results of the contaminant transport simulation show that As, Cd and Cr will reach the water table after 162 years, 104 years and 201 years, respectively. Movement of these contaminants with ground water will be governed by their respective R_d values in the subsoil below the water table. The R_d values for Durango tailings leachate and Soil B in Table 3 along with Equation (1) give retardation factors for these contaminants of:

As	$R_f = 2.70$
Cd	$R_f = 2.70$
Cr	$R_f = 4.41$

Travel time for each of these contaminants from the edge of the pit to a 100 m boundary is found by multiplying the retardation factor by the ground-water travel time (328 years). Those travel times are:

As	887 years
Cd	887 years
Cr	1446 years

The total travel times for each contaminant is the sum of the time required to reach the water table plus the time need to move from the edge of the pit to the 100 m boundary. Total travel times for the selected contaminants in Durango leachate are:

As	1049 years
Cd	991 years
Cr	1647 years

From this analysis we find that As and Cr are expected to satisfy the established objectives, but Cd falls just short of meeting the criteria. As stated previously, these results are based on the assumption that dilution dispersion and diffusion effects are negligible. If an actual site evaluation reveals that a few contaminants, such as As and Cd for an unlined site, are borderline cases (as far as satisfying the site specific criteria is concerned), then the transport model used must include the effects of dilution, dispersion and diffusion.

4.3.2.2 Acidic Tailings

Prediction of contaminant transport through the liner requires (in addition to the data needed for water flow prediction) the R_d value for the pH and two R_d values for each contaminant. The two contaminant R_d values describe the movement of a specific contaminant in both unneutralized and neutralized leachate. R_d values for selected contaminants in the Shiprock tailings leachate are given in Table 3. Computer simulation results show that the buffering capacity of soil A is very effective in delaying the arrival of unneutralized leachate at the water table. Movement of the acid leachate front ($\text{pH} < 7.7$) from the bottom of the pit to the water table is shown in Figure 12. Without a liner, the unneutralized leachate took 61 years to reach the water table, versus 227 years with the liner.

The retardation factor for the acid leachate front ($\text{pH} < 7.7$) in the soil below the water table is 1.34; thus another 440 years are needed (in addition to the 227 years to reach the water table) before the leachate would reach a 100 m site boundary. In all it would be 667 years. Thus, these three contaminants do not satisfy the 1000 year criteria.

In an actual site evaluation, the leachate leaving the tailings pit would not remain acidic for as long as is assumed in this demonstration. The liner and the subsoil below the water table would probably neutralize much more leachate (because the acidity decreases as the tailings are leached) and travel times for As, Cd and Cr would be increased significantly.

The radionuclides Pb, Ra, U and Th are retarded significantly in the acidic leachate through both the liner and the subsoil below the water table. Travel times for Pb and Ra from the top of the clay liner to the water table

FIGURE 12. Movement of the Acid Leachate ($\text{pH} < 7.7$) from the Bottom of the Tailings Pit to the Water Table. (The dashed line indicates an unlined pit and the dotted line, a lined pit.)

are approximately 280 and 760 years, respectively. Travel time from the edge of the pit to the 100 m boundary is found by multiplying their respective retardation factors (under saturated conditions in Soil B) times the travel time for water. Total travel times to the boundary for Pb and Ra are about 1400 years and 12,000 years, respectively, which satisfies the design objectives established for this demonstration.

Because U and Th in unneutralized leachate move faster than the acid front through the clay liner (but much slower than the acid front if they are in neutralized leachate), both these elements would move in a "roll front" with the same low velocity as the acid front as long as they are in the clay liner. Travel times for U and Th to the water table are 260 and 270 years,

respectively. Total travel times to the site boundary at 100 m are about 1400 years and 1800 years, respectively, for U and Th, which also satisfy the EPA criteria proposed in 1981.

The presence of the liner delays the arrival of Pb and U at the 100 m site boundary by approximately 200 years. Thorium arrival is delayed by 100 years. Since these elements have already met the EPA criteria proposed in 1981 for the unlined pit, the delay caused by the liner's neutralization capacity is added protection against their release but is not needed to meet the criteria. For As, Cd and Cr, however, the delay in arrival time may be crucial. The liner's buffering capacity is capable of neutralizing just over one pore volume of the worst case tailings leachate (shown in Table 2 for Shiprock tailings leachate). Because the total acidity of the leachate decreases rapidly at the end of the first pore volume, the acid front will require significantly more time to reach the 100 m boundary than predicted by this analysis (for which constant acidity is assumed). Contaminants such as As, Cr and Cd that move with the acid leachate front will also take longer to reach the boundary.

In summary, a procedure has been demonstrated for determining the need and type of liner at a disposed site with groundwater contamination potential. The procedure requires characterization of the site hydrology, tailings contaminant leach rates, contaminant concentrations in leachate and interactions between leachate and each layer of soil. The decision tree procedure demonstrated here offers the users of this document a method to evaluate the disposal site and compare its performance with site specific objectives.

5.0 SUMMARY

Liners and foundation soils under disposal pits for uranium mill tailings from inactive sites are the major engineered barriers for preventing contaminant release to ground and surface water. The purpose of this report is to provide a logical sequence of action, in the form of a decision tree, which may be followed for sites with potential groundwater problems, to show whether or not a selected tailings disposal design satisfies established objectives for that site in terms of subsurface contaminant release.

The decision tree is based on the capability of hydrologic and mass transport models to predict the movement of water and contaminants with time. Predictions of contaminant movement can then be compared with the design objectives for contaminant release to determine whether or not a site meets those standards. If a site disposal design without a liner meets the standards, no liner is required. If an unlined site fails to satisfy the criteria for contaminant release, then either a design that includes a liner must be evaluated or new objectives must be established. If the addition of a liner to the design fails to satisfy the objectives, a new liner, disposal site design, or an alternative site must be chosen.

Evaluation of a site begins with characterization, which includes measuring moisture transport properties of cover layers, tailings, candidate liners and subsurface soils or near surface aquifers. Saturated and unsaturated hydraulic conductivities, moisture retention curves and layer depths and locations are needed for each layer of material present. Contaminant leach rates and concentrations in leachate from the tailings are needed. Transport properties (buffering capacities and retardation factors) for contaminants are required for all layers through which the leachate may migrate.

The decision tree procedure was demonstrated for a generic site in the western U.S. Several of the hydrologic and transport models that might be used and their data requirements are discussed. A design for tailings disposal in a pit is selected and evaluated for two different types of tailings. The tailings selected are an alkaline processed tailings and an acid processed tailings. Movement of contaminants from both tailings are analyzed for both a lined and an unlined tailings pit. These results are compared with the

objectives established for this demonstration. Because of the generic nature of the decision tree demonstration, some simplifications were made to avoid complications that might detract from the point of this document: to provide a guide for evaluating a site with potential groundwater problems to determine if a liner is needed. At points where simplifying assumptions have been made, the assumptions are spelled out and instructions are given as to how a specific site might be treated differently.

In the demonstration given here, a soil liner ($K \approx 10^{-9}$ m/s) was utilized in a computer simulation of water movement and contaminant transport from a uranium mill tailings disposal site. The movement of water from the pit to the site boundary was not predicted to be changed significantly by the presence of a liner. However, the liner was found to be significant in delaying contaminant release. By neutralizing acidic leachate and causing contaminants to precipitate, the acid leachate travel time to a 100 m boundary was increased by one third. The travel times for contaminants in alkaline leachate were increased by <10% with the addition of a liner to the pit. To effectively reduce contaminant release to subsurface and surface water, cover layers should be carefully designed to reduce infiltration of water from above and soil liner material should be used to attenuate contaminant concentrations in leachate.

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APPENDIX

DATA REQUIREMENTS FOR THE DECISION TREE

APPENDIX

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CALCULATION OF INFILTRATION THROUGH THE COVER USING THE UNSATURATED FLOW MODEL UNSAT1D (Bond et al. 1982)

This model was used to calculate infiltration through the cover layers to the tailings pile. Monthly summaries of weather data from Grand Junction, Colorado for 1979 were provided by the National Oceanic and Atmospheric Administration (NOAA). Potential evaporation was calculated using temperature, humidity, wind speed, sunshine, radiation and vegetative cover data. The precipitation calculated potential evaporation data were combined with data on root uptake capability in the model to predict water infiltration by unsaturated flow through the cover layers.

Saturated hydraulic conductivity values assumed for soil A (liner) and soil B (sandy material) were 4.4×10^{-9} and 1.2×10^{-6} m/s, respectively. Saturated volumetric water contents (porosity) for soil A and soil B were 0.40 and $0.44 \text{ m}^3/\text{m}^3$, respectively. Soil moisture retention curves for these soils were assumed to be the same as soil for soil B and shale (packed to a bulk density of $1.5 \text{ mg}/\text{m}^3$) for soil A in Bond et al. (1982). Unsaturated soil hydraulic conductivities were calculated from soil moisture retention data using HYDRAK and fit to a polynomial equation using POLYFIT [HYDRAK and POLYFIT are subroutines of UNSAT1D (Bond et al. 1982)].

CALCULATION OF WATER MOVEMENT THROUGH TAILINGS, LINER AND SOIL TO THE WATER TABLE

The water flow model TRUST (Reisenauer et al. 1982) was used to predict the unsaturated water movement through the tailings liner and soil below the pit to the water table. The infiltration from UNSAT1D was used as the boundary condition at the overburden (soil A)-tailings interface. The lower boundary condition was the existence of a water table (saturation) at 2.4 m below the tailings pile. The initial moisture condition of each soil and the tailings was essentially air dry. Unsaturated hydraulic conductivities of soil A for

the liner and soil B below the pit were unchanged from the UNSAT1D run. Saturated conductivity and water content for the tailings were assumed to be 2.2×10^{-6} m/s and $0.44 \text{ m}^3/\text{m}^3$. Unsaturated hydraulic conductivity values for the tailings were taken from Nelson et al. (1980).

Two cases were selected for analysis of water flow using TRUST. These cases are shown graphically in Figure 6 and 7 and differ only by the absence of a liner in Case 1 (Figure 6) and presence of a liner (Figure 7) in Case 2.

CONTAMINANT TRANSPORT

Movement of water as a function of time and distance in one dimension is predicted by TRUST. Prediction of contaminant migration requires the additional data given in Table 3. The TRUST subroutine MLTRAN [a modified version of MILTVL (Nelson, Reisenauer and Gee 1980)] is then used to predict contaminant locations as a function of time and distance traveled. If the two or three dimensional versions of TRUST are used, MLTRAN would predict contaminant transport as a function of time and distance in two or three dimensions.

CONTAMINANT RETARDATION DATA

Below are the raw data used to calculate the distribution ratio values for selected contaminants in Durango and Shiprock tailings leachates with soil A.

Batch Test Results for Durango Tailings Leachate

	<u>Initial Values</u>	<u>Final Values</u>	<u>Rd (ml/g)^(a)</u>
pH	9.7	9.4	
As	1.2 ppm	0.7	3.5
Cd	0.01 ppm	<0.01	≈1
Cr	0.07 ppm	0.04	3.8

(a) The solution volume (v) to solid weight (w) ratio required for calculation of an Rd using equation (2) was $v/w = 5.0$.

Shiprock Tailings Leachate Batch Test Results

	<u>Initial Values</u>	<u>Final Values</u>	<u>Rd (ml/g)^(a)</u>
pH	2.1	7.7	
As	3.8	0.2	90
Cd	4.7	0.05	465
Cr	1.9	<0.02	>470
Pb	5.0	0.2	120

(a) The solution volume (v) to solid weight (w) ratio required for calculation of an Rd using equation (2) was $v/w = 5.0$.

LINER HYDRAULIC CONDUCTIVITY FOR DURANGO TAILINGS LEACHATE

A column containing soil A was packed to a bulk density of 1.80 mg/m^3 and used to measure the hydraulic conductivity with synthetic Durango tailings leachate. After 4 months, only 0.70 liner pore volumes had passed through the column at an almost constant rate. Because of the high bulk density, the initial permeability was low ($K = 1.2 \times 10^{-11} \text{ m/s}$). Over the 4 month period, however, K varied from 1.1×10^{-11} to $3.0 \times 10^{-11} \text{ m/s}$. In a similar soil from Clive having a clay content of 54%, a bulk density of 1.54 mg/m^3 gave an initial value of $K = 6.1 \times 10^{-10} \text{ m/s}$ and ranged from 1.5×10^{-9} to $3.2 \times 10^{-9} \text{ m/s}$ for the next 2 weeks. During this time, 3.5 pore volumes of Durango tailings leachate passed through the column. The hydraulic conductivity of the Clive soils with Durango leachate is very dependent on the bulk density but relatively unchanged with time.

LINER HYDRAULIC CONDUCTIVITY FOR SHIPROCK TAILINGS LEACHATE

One column of soil A (bulk density = 1.84 mg/m^3) was tested with synthetic leachate similar to Shiprock tailings leachate. Approximately 0.1 pore volumes passed through the column before it plugged up completely. A similar soil with higher clay content and a bulk density of 1.55 mg/m^3 was used for another test with synthetic Shiprock leachate. In this test, the initial K value was $8.5 \times 10^{-10} \text{ m/s}$; however, only 0.25 pore volumes of leachate passed through the column before K dropped to $3 \times 10^{-13} \text{ m/s}$ and plugged up.

The two Clive surface soils tested were high in calcium carbonate content (27 and 55% CaCO_3). The acidic leachate from Shiprock tailings is neutralized

very well by these soils and much of the contamination is removed by precipitation. After contacting these soils with Shiprock leachate in batch tests, gypsum was found (by X-ray diffraction) to have formed. The data indicate that high sulfate, acidic leachates plug up these liner materials (surface soils) by precipitation of gypsum and other minerals to form a liner with extremely low permeability. If the decrease in permeability with time had been included in the TRUST model, the tailings pit would have begun to fill with water and likely have escaped through the pit walls to ground water or to the surface. Modeling of this phenomenon would require the two dimensional version of TRUST.

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