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Liner Evaluation for Uranium Mill Tailings: Final Report

**Compiled by
J. L. Buelt**

September 1983

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LINER EVALUATION FOR URANIUM MILL TAILINGS:
FINAL REPORT

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SUMMARY

The Liner Evaluation for Uranium Mill Tailings Program was conducted to evaluate the need for and performance of prospective lining materials for the long-term management of inactive uranium mill tailings piles. On the basis of program results, two materials have been identified:

- natural foundation soil amended with 10% sodium bentonite
- catalytic airblown asphalt membrane.

The study showed that, for most situations, calcareous soils typical of Western U.S. sites adequately buffer tailings leachates and prevent groundwater contamination without additional liner materials or amendments. Although mathematical modeling of disposal sites is recommended on a site-specific basis, there appears to be no reason to expect significant infiltration through the cover for most Western sites. The major water source through the tailings would be groundwater movement at sites with shallow groundwater tables. Even so, column leaching studies showed that contaminant source terms were reduced to near maximum contaminant levels (MCL's) for drinking water within one or two pore volumes; thus, a limited source term for groundwater contamination exists. At sites where significant groundwater movement or infiltration is expected and the tailings leachates are alkaline, however, the sodium bentonite or asphalt membrane may be necessary.

The Liner Evaluation Program conducted by Pacific Northwest Laboratory (PNL) can be divided into five major categories:

- tailings characterization studies
- preliminary studies
- liner evaluations by laboratory tests
- liner evaluations by field tests
- decision tree development.

The tailings characterization studies included batch and column leaching studies to determine the contaminant source term in the leachate for the laboratory studies. The preliminary studies identified and evaluated various liner materials and selected eight of the most promising for study. The laboratory liner tests screened the eight materials to identify an asphalt and a clay liner for further study and subjected these materials to controlled evaluations to quantify their expected performance lifetime. The asphalt and clay liners were also subjected to evaluations in the field. During the decision tree development, a logical approach was developed through the use of mathematical models to identify the need for and type of liner at a disposal site.

TAILINGS CHARACTERIZATION STUDIES

The objective of the tailings characterization studies was to identify tailings leachate concentrations for liner exposure and interaction studies in the laboratory. Various tailings samples from Shiprock, New Mexico, Salt Lake City, Utah; and Durango, Colorado, were collected and subjected to 5:1 and 100:1 batch leach tests. Because the leach tests were considered inadequate to identify contaminant concentrations as a function of time, column leaching studies were performed on the most highly contaminated batch samples. The columns produced tailings leachate that indicated a limited contaminant source term. Within two pore volumes, contaminant concentrations were near MCL's for drinking waters. These data are interpreted to mean that only contaminants from the first two pore volumes of leachate need be considered when determining liner requirements.

PRELIMINARY STUDIES

At the beginning of the program, various liners were evaluated by referring to published literature. The types of liners included clays, soil sealants, synthetic geomembranes, asphalts, and concretes. The following materials were identified for laboratory screening:

- catalytic airblown asphalt
- hydraulic asphalt concrete

- asphalt-rubber membrane
- chlorosulfonated polyethylene
- natural soil
- soil amended with sodium bentonite
- soil amended with Saline Seal-100^(a)
- soil amended with GSR-60.^(a)

LABORATORY TESTS

Each of the materials was subjected to extreme conditions in 0.6-m diameter exposure columns to duplicate field conditions as closely as possible while subjecting them to increased temperature, acidity, oxygen concentrations, and hydrostatic pressure to accelerate aging reactions. Based on the indicated stability, effectiveness as a leachate barrier, and cost, catalytic airblown asphalt and natural soil amended with sodium bentonite were selected for further study.

The catalytic airblown asphalt was subjected to a series of aging tests in the exposure columns under various temperatures and levels of oxygen and acidity. Aging reaction products were analyzed by infrared spectrometry and results indicated that the asphalt was extremely stable. An aging period equivalent to 7 years was measured, yet the extent and penetration of reaction products was so minute that 1000-year liner performance can be expected to be exceeded under uranium mill tailings impoundment conditions.

Natural soils from Clive, Utah, and soils amended with natural sodium bentonite were subjected to material characterization, batch interaction, and column interaction studies. Results showed that the high CaCO_3 content, typical of Western sites, provided an adequate buffering capacity and

(a) Saline-Seal 100 and GSR-60 are registered trademarks of the American Colloid Company.

caused contaminant coprecipitation in the leachates even without bentonite amendments. This behavior was demonstrated in the column interaction studies, which showed that calcareous soils in contact with acidic leachates become plugged due to coprecipitation, producing permeabilities between 10^{-9} and 10^{-10} cm/s.

Natural soils in contact with alkaline leachates maintained relatively high initial permeability and demonstrated negligible sorptive or buffering characteristics. When amended with sodium bentonite, however, the clay liner permeability decreased with time while in contact with alkaline leachates. Reductions of cationic contaminant concentrations due to sorption were also observed.

FIELD TESTS

Catalytic airblown asphalt and sodium bentonite amended soil were also evaluated in the field. The materials were placed in a 1.8-m deep pit in a Grand Junction, Colorado, tailings pile in 1981. Each liner occupied half the pit over a 0.3-m thick foundation soil, which was instrumented with moisture blocks and moisture extraction cups to obtain readings and samples of moisture migrating through the liners. After a 2-year exposure, the asphalt liner had retained its flexible characteristics and shows little degradation when compared with a virgin sample.

Low moisture saturation levels were maintained in the foundation soil because of the impervious asphalt liner. Insufficient asphalt concentrations occurred over a one-square-meter area, which contained small holes (approximately 1 cm in diameter) that allowed the foundation soil directly beneath it to become saturated. This experience exemplifies the need for proper engineering and inspection practices when applying an asphalt membrane.

The clay liner did not keep the foundation soil dry for more than three months because it is much more permeable and the leachates were not acidic (pH = 7 to 8). The beneficial characteristics of this liner are

associated with the sorptive properties of the sodium bentonite in the clay. Comparisons of moisture above and below the clay liner showed that contaminant concentrations were reduced as the leachate passed through the liner. Analysis of clay liner cores after the 2-year exposure verified that contaminants are not mobile through this material.

DECISION TREE DEVELOPMENT

With the permeabilities, buffering, and sorptive characteristics of the two liner materials identified, a logical approach was developed to combine these data with hydrologic models and contaminant source terms from the tailings. This approach, called a decision tree, is a step-by-step sequence that can be used to identify the need for and type of liner at a particular disposal site. Models such as the TRUST code and UNSATID can be used to determine the long-term (1000-year) behavior of the impoundment as it pertains to groundwater quality.

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INTRODUCTION

A number of inactive uranium tailings piles have accumulated from milling operations throughout the United States. In 1978, Congress passed Public Law 95-604, which requires disposal of the 24 sites that are the responsibility of the U.S. Department of Energy (DOE). In response, DOE created the Uranium Mill Tailings Remedial Action Program (UMTRAP); one of the program activities was to develop and evaluate technologies for disposal of the piles in conformance with regulatory criteria established by the U.S. Environmental Protection Agency (EPA). The disposal technologies required, among other things, a program to evaluate the need for and type of liners for long-term (1000-year) protection of the groundwater. For this reason, UMTRAP created the Liner Evaluation for Uranium Mill Tailings Program conducted by Pacific Northwest Laboratory (PNL). This final report is a compilation of all relevant results and conclusions of the PNL liner program.

The PNL liner evaluation program was established late in FY-1980 and was completed in FY-1983. It consisted of five major areas of activity:

- tailings characterization studies
- preliminary liner studies
- liner evaluations by laboratory tests
- liner evaluations by field tests
- decision tree development.

The selected approach was to determine the long-term performance of two of the most promising liner materials identified through a preliminary literature survey and laboratory screening tests. Evaluation of the two liner materials, a natural soil amended with sodium bentonite and an asphalt membrane, included interaction with various tailings leachate compositions identified in the tailings characterization studies and assessment after a 2-year field exposure. The study data can be used in the decision tree, which is a sequence developed to determine the long-term need for and performance of a liner under uranium tailings impoundment conditions.

CONCLUSIONS

Through some of the findings of the PNL liner program and other UMTRAP programs, it became apparent that the natural retarding properties of disposal site soils are, in most cases, enough to prevent groundwater contamination from mill tailings. Calcareous soils, typical of Western U.S. sites, interact with acidic leachate and cause contaminant coprecipitation as the leachate passes through the foundation soil/tailings interface. Where groundwater movement or infiltration through alkaline tailings exists, however, sodium bentonite amended soils or a catalytic airblown asphalt membrane may be required if the groundwater quality objectives established for the disposal site are exceeded. Laboratory evaluations and field studies identified these two materials as the most promising.

In addition to the above findings, the following major conclusions have been derived from the PNL program activities:

- Contaminant releases from tailings as leachate passes through them are limited. For example, the column leaching studies showed that concentrations of all radionuclides and chemical contaminants were reduced to near or below maximum contaminant levels (MCL's) for drinking water (Environmental Protection Agency 40 CFR 141) within one or two tailings pore volumes.
- Sodium bentonite amended soils and catalytic airblown asphalt proved to be the most effective materials as leachate and contaminant barriers. They were also among the least expensive of the eight materials selected for testing during the preliminary studies.
- Accelerated aging tests successfully exposed the asphalt membrane to the equivalent of a 7-year exposure over a 3-month period. Reaction rates were extremely slow and penetration of reaction products was limited to 0.5% of the total membrane thickness.

The data indicate that a performance period greater than 1000 years can be expected when this material is exposed to uranium mill tailings impoundment conditions.

- Analysis of the field-exposed asphalt membrane demonstrated similar penetration of reaction products. After a 2-year exposure, the liner retained its pliable characteristics; however, due to inadequate membrane thickness in a small area, some leakage was noted through 1-cm diameter holes. This experience exemplifies the need for proper engineering and inspection practices.
- The buffering capacity of a highly calcareous soil from Clive, Utah, once considered as a disposal site and chosen for the PNL clay/soil evaluations, was 1100 meq/100 g of soil (i.e., 100 g of soil without amendments will neutralize up to 11,000 liters of acidic leachate from pH = 2 to pH = 4.5, which is more than adequate to prevent groundwater contamination).
- Coprecipitation of contaminants from acidic leachates in contact with calcareous soils tends to plug the pores and reduces permeabilities to 10^{-10} cm/s.
- Calcareous soils in contact with alkaline leachates exhibit no sorptive properties and maintain their relatively high permeabilities unless amended with 10% sodium bentonite.
- Although the clay liner installed in the field in contact with neutral pH leachates did not keep its foundation soil dry for more than 3 months, it did reduce contaminant concentrations effectively as the leachate passed through the liner.
- The decision tree developed during this program could be useful in evaluating the need for and type of liner at specific disposal sites. This analytical tool considers site-specific conditions and evaluates the long-term groundwater quality through data compiled in the PNL program and from existing hydrologic models.

TAILINGS CHARACTERIZATION STUDY

The objective of the characterization study of the Liner Evaluation Program was to determine the chemical properties of tailings at 3 of the 24 DOE tailings sites. In turn, the chemical contents of the tailings were used to determine the conditions under which the effectiveness of the liners would be tested.

Tailings samples were taken from the Salt Lake City, Utah; Durango, Colorado; and Shiprock, New Mexico, sites, which were sampled at the locations indicated in Figures 1, 2, and 3. Each sample was composited over a depth of about 1.8 m. At the time of sampling, a wide range of textures was noted among locations in a single pile, apparently the result of how the slimes and sands settled in the original tailings pond. At Salt Lake City, samples taken for moisture determinations indicated that moisture content correlated with increased fineness of texture.

FIGURE 1. Sample Sites on the Salt Lake City Vitro Tailings Pile

FIGURE 2. Sample Sites on the Durango Tailings Pile

FIGURE 3. Sample Sites on the Shiprock Tailings Pile

The tailings samples were sealed in buckets and returned to the laboratory, where they were analyzed for cations, anions, hydrocarbons, and radionuclides. Extracts for cation and anion analyses were obtained by shaking tailings with water in a 100 to 1 (by weight) concentration for about 2 hours. Cations were analyzed by inductively coupled argon plasma techniques, the anions by ion chromatography, and the radionuclides by gamma scan. Hydrocarbon concentrations were assumed to be negligible. Analytical results are shown in Table 1. Although these methods detect and/or quantify a number of elements and ions, only those of special interest to this study are included in the table.

The 100 to 1 leachate samples were used to determine the total quantity of leachable material. To assess the composition of tailings leachate in contact with a liner, the batch leaching technique is adequate to screen a large number of samples but is inadequate to quantify contaminant releases over time. As indicated in recent studies (Relyea, Martin and Colclough 1982), batch leach tests generally underestimate contaminant concentrations that are initially leached from tailings. In fact, batch leaching tests yield little, if any, information on contaminant release rates. (See Table 2 for a comparison of leachate composition from 5 to 1 batch tests and column tests.) Therefore, column leaching tests were conducted to characterize contaminant release rates and concentrations from tailing samples.

Results from column leach tests on tailings samples have shown that the highest concentrations of contaminants are released in the first or second pore volumes of leachate. As shown in Figure 4, concentrations of radionuclides ^{238}U , ^{230}Th and ^{226}Ra decreased by at least an order of magnitude after one pore volume. However, radioactive element concentrations after two pore volumes were still observed to be above the EPA proposed MCL's for all three tailings samples (USEPA 40 CFR 141). Contaminant concentrations of Al, As, Cd, Cr, Cu, Fe, Mg, Mn, Mo, Ni, P, SO_4 and Zn were also highest in the first increment of leachate from the three tailings samples tested and decreased by an order of magnitude or more at the end of one pore volume.

TABLE 1. Analytical Results From Tailings at Salt Lake, Durango and Shiprock Sites Using 100:1 Lechate-to-Sample Ratio

Salt Lake	Sample Number					
	1	2	3	4	5	6
pH	2.9	7.4	8.0		6.7	5.5
Al (ppm in tailings sample)	109	26	46	11	14	19
Ca (ppm)	24,200	14,000	8,290	26,000	36,200	25,500
Fe (ppm)		30	23	500	2	3
Mg (ppm)	62	88	194	400	230	256
Na (ppm)	162	290	235	1,800	1,330	660
Si (ppm)	17	238	305	566	153	87
SO ₄ (ppm) (a)	2,400	1,670	1,670	8,140	2,720	2,230
NO ₃ (ppm)	<1	26	87	<1	383	310
Cl (ppm)	6.4	12	14	1,333	79	15
²¹⁰ Pb (dpm/g)	2,430		1,370		1,970	
²³⁸ U and ²³⁴ Th (dpm/g)	90		190		49	
²³⁰ Th (dpm/g)	960		620		460	
²³⁵ U (dpm/g)	<17		10		29	
²²⁶ Ra (dpm/g)	2,270		1,240		2,000	

Durango	Sample Number										
	1	2	3	4	6	7	8	9	10	11	
pH	9.9	10.0	9.8	10.0	9.4	9.5	7.1	7.5	9.6	7.0	
Al (ppm in tailings sample)	32	11	11	17	63	162	6	16	11	<3	
Ca (ppm)	1,170	1,140	800	1,060	375	989	2,170	600	860	174	
Fe (ppm)	4.1	--	2	1	23	35	4	6	--	2	
Mg (ppm)	132	100	18	86	37	68	--	11	136	--	
Na (ppm)	230	110	116	377	75	112	32	45	368	32	
Si (ppm)	106	60	130	73	343	500	90	100	198	38	
CO ₃ (ppm- assumed from inorganic carbon analysis)	3,500	3,060	1,600	2,500	1,590	2,000	280	365	3,100	280	
SO ₄	low	low	low	low	--	--	--	--	--	--	
²¹⁰ Pb		1,500			1,530					995	
²³⁸ U and ²³⁴ Th		575			275					154	
²³⁰ Th		2,840			2,035					1,520	
²³⁵ U		27			<14					12	
²²⁶ Ra		2,300			1,940					1,360	

Shiprock	Sample Number										
	1A	1B	2	3	4	5	6	8	9	10	11
pH	7.0	4.1	4.1	4.6	6.3	4.4	4.4	4.6	4.2	4.8	5.1
Al (ppm in tailings sample)	21	97	367	31	8	99	187	<3	173	7	3
Ca (ppm)	7,790	6,300	15,500	7,220	13,500	39,700	44,600	8,950	34,400	7,290	7,230
Fe (ppm)	7	29	35	3	3	1	6	1	2	2	
Mg (ppm)	182	160	320	61	210	466	511	37	183	29	58
Na (ppm)	180	340	260	83	232	518	523	34	139	39	144
Si (ppm)	185	345	32	51	191	61	69	69	52	37	55
SO ₄ (ppm)	19,000	17,600	45,200	20,900	33,300	101,300	112,500	22,100	88,400	20,000	18,800
NO ₃ (ppm)	33.5	13.0	10.0	4.6	21.2	37.2	34.0	6.9	2.0	2.8	15.4
Cl (ppm)	4.0	4.3	3.0	3.2	4.7	9.4	17.5	3.1	13.2	4.0	3.0
²¹⁰ Pb (dpm/g)	605		1,170			3,700					
²³⁸ U and ²³⁴ Th (dpm/g)	25		33			<23					
²³⁰ Th (dpm/g)	340		370			1,900					
²³⁵ U (dpm/g)	<8		10			<27					
²²⁶ Ra (dpm/g)	510		1,100			4,025					

(a) Performed with 1:1 leachate-to-sample ratio. Concentration limited by solubility product.

TABLE 2. Comparison of Column and Batch Leach Tests: Chemical Composition of Tailings Leachates (Concentrations in mg/L)

	Salt Lake		Shiprock		Durango	
	5:1 Batch ^(a)	Column ^(b)	5:1 Batch	Column	5:1 Batch	Column
Al	4.0	1200.0	66.0	2700.0	0.6	11.0
Ca	600.0	550.0	560.0	530.0	1.1	7.2
Cd	0.05	2.6	0.1	5.3	<0.01	0.15
Cu	1.9	48.0	3.5	130.0	ND ^(c)	ND
Fe	6.7	1700.0	2.4	160.0	ND	0.17
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
Zn	2.2	98.0	2.9	120.0	ND	0.12
pH	3.0	2.0	3.1	2.3	9.3	9.5

(a) The 5:1 batch tests utilize 5 cm³ of distilled water for each gram of tailings.

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

(c) ND = Not Detected in the sample.

Concentrations of As, Cd, and Cr in leachate from acidic Salt Lake and Shiprock tailings (shown in Figure 5) were near or below the EPA proposed MCL levels after two pore volumes. Similarly, leachate concentrations of As, Cd, and Cr from alkaline Durango tailings decreased to near or below MCL after one or two pore volumes. Ba and Pb were below MCL in all samples.

In conclusion, although the batch leaching studies agreed well with the results of Markos and Bush (1981a,b), they did not portray release rates accurately. The column leaching studies demonstrated that contaminant concentrations are markedly reduced after the first two tailings pore volumes of leachate.

FIGURE 4. Concentrations of Radioactive Elements ^{230}Th , ^{238}U , and ^{226}Ra in Tailings Leachates. Initial concentrations (first effluent) are labeled 0. Concentrations at the end of 1 and 2 pore volumes are labeled 1 and 2, respectively.

FIGURE 5. Concentrations of Nonradioactive Hazardous Elements in Tailings Leachates. Initial concentrations (first effluent) are labeled 0. Concentrations at the end of 1 pore volume and 2-1/2 pore volumes are labeled 1 and 2.5, respectively. An arrow on the bar denotes a measured value below detection.

PRELIMINARY STUDIES

The preliminary studies were conducted to select eight of the most promising liner materials from those considered for laboratory testing. The studies included a review of published literature and information obtained from industrial contacts. Clays, soil treatments, asphalt membranes, asphalt concretes, grouts and concretes, and polymeric membranes were considered; and the materials were evaluated for long-term effectiveness as a moisture and/or contaminant barrier with cost effectiveness in mind.

CLAYS

Various clay liners have been installed beneath impoundments containing waste materials. These liners include untreated natural clay minerals and clays artificially saturated with sodium to resist loss of sealing properties when leached by wastes of high ionic strength (Fuller 1981; Griffin and Shimp 1978; Haxo 1976a; Haxo and White 1976b; Lubina, Hovater and McCready 1979; Williams 1978; Shultz and Miklas 1980).

The chemical properties of the waste and the methods used to fill the impoundment influence the choice and design of the liner system. If exposed to natural weathering cycles such as wet/dry and freeze/thaw, a clay liner can lose much of the sealing ability it had when installed (Rollins and Dylla 1970). However, if the clay liner is initially hydrated with fresh water and covered to prevent drying, it can form an effective seal.

The sealing property of clay results from its ability to expand when wet. This expansion decreases the pore space of the soil and its permeability. Clay minerals called smectite or montmorillonite are generally responsible for this sealing property. Other clay minerals such as illite and kaolinite also expand when wet, but not as much as montmorillonite.

The ions present on the exchange sites of the clays also influence the amount of shrinking and swelling. In the presence of solutions containing

high concentrations of dissolved salts, Na in prehydrated clay may be replaced by other ions such as Ca and Mg, causing the clay to shrink and lose some of its effectiveness as a sealant. Na-saturated montmorillonite, illite, and kaolinite swell more than the same clays saturated with Ca. Clays subject to less swelling have been used as sealants on a limited basis. These clays are affected less by increased concentrations of Mg or Ca, and the shrinkage caused by drying may also be less. The selection of clay minerals to be used in a liner will depend on availability at specific site locations as well as the characteristics of the leachate within the mill tailings pile.

A special form of montmorillonite, Saline Seal 100, is reported to be resistant to attack by high concentrations of ionic contaminants (Hughes 1974); and this material was selected for the laboratory tests. The Saline Seal 100 clay was mixed with natural soil to determine optimum composition for a clay liner. In addition, natural soils from typical sites and sodium bentonite amendments were studied to compare sealing properties with Saline Seal 100.

One significant advantage of natural clay minerals over geomembrane materials is their low cost (U.S. NRC 1979), especially if they can be obtained close to the disposal site. Another advantage is their self-healing ability. If the liner fails because of minor breaks, the clay will seal itself by filling up the breach in the liner with particles over a period of time. This self-healing ability eliminates problems that can arise if ground settling or other events cause damage to the liner (Hughes 1974).

SOIL SEALANT MATERIALS

Studies have shown that permeability can be reduced by the application of various chemicals that react with the soil. However, most soil sealants have been reported to be affected by freezing/thawing, wetting/drying, reactions with leachates, and leaching of the sealing agent by the waste liquid, as Stewart (1978) discusses below.

"Most of these materials (sealants) consist of a blend of a high-molecular-weight linear polymer and a crosslinked, swellable polymer of approximately the same molecular weight. The linear polymer's many sorptive sites allow it to sorb to the soil and form a flexible network. The cross-linked polymer is extrudable and can conform to permeability channels in the soil without loss of integrity. In use, the polymer system helps to form a stabilized soil surface of extremely low permeability.

"Formulation of the polymeric soil sealant system depends on its use, and in most instances, it is considered to be proprietary by the marketing company. When used in unfilled impoundments, the polymer is mixed in a low-pH water/acid solution and then sprayed on the earthen surface as a low-viscosity slurry. The low-pH condition allows the polymer to penetrate the surface. Upon subsequent exposure to water, the water-swellable portion swells and becomes locked in place. The linear polymer, being sorptive in this state, attaches to the soil to complete formation of the stable, impermeable surface."

No actual field data are available, but Stewart also discusses sealant limitations, including polymer shrinkage, that affect the seal. Because of these limitations, the sealants identified by Stewart (1978) in Table 3 did not receive further consideration as long-term moisture barriers.

ASPHALTS

The low water permeability of asphaltic compositions was known to ancient peoples, and the long-term stability of these compositions is evidenced by the survival of several early asphaltic structures (The Asphalt Institute 1976). They are versatile in application and can be alloyed with various materials, creating compounds to meet a range of situations. Asphalt-based compounds can be formulated to resist temperature extremes, to provide water permeability less than 10^{-9} cm/s (Hickle 1976), and for a variety of application methods. Asphalts resist most oxidizing acids, except nitric, and attacks from salts and alkalies up to concentrations of 30% in solution. However, these compounds are

- oxidized by exposure to ultraviolet radiation
- sometimes attacked by microbes if not protected by biocides

- susceptible to subgrade displacement due to poor compaction, freeze/thaw volume changes, and subsidence
- soluble in solutions >5% hydrocarbons.

Should further characterization of the tailings leachate reveal significant quantities of the organics used in the solvent extraction process (kerosene, isodecanol, and alanine) (U.S. NRC 1980), asphaltic liners can be protected by the application of a final tar seal coat.

TABLE 3. Representative Soil Sealants (Stewart 1978)

Sealant	Application	Remarks
Cationic Asphalt Emulsion	Farm Ponds	Requires ~19,000 L/4,047 m ² (5000 gal/acre) dispersed in water.
Oil-Soluble Polymers in Diesel Fuel	Fresh Water	Injected beneath surface of water where seepage was occurring.
Sodium Tetraphosphate	Sulfite Liquor Storage	Dispersant distributed in 15.2-cm (6-in.) layer of soil at 2.3 kg/9 m ² (5 lb/100 ft ²). Careful compaction rendered soil impervious.
Sodium Carbonate	Canals	Wet-dry cycles disrupt water barrier. Used 183 g (0.4 lb) of reagent per 0.84 m ² (yd ²) of soil.
Lignin Derivatives Gelled Alum	Desalination Byproduct Brine	1% lignin cost \$3400 per 4047 m ² (acre).
Carboxymethyl Cellulose (CMC) with Alum	Desalination Byproduct Brine	0.2% CMC cost \$2250 per 4047 m ² (acre).
Petroleum Emulsions	Desalination Byproduct Brine	4% additive cost \$4400 per 4047 m ² (acre).
Attapulgite Clay	Desalination Byproduct Brine	2% Zeogel cost \$1000 per 4047 m ² (acre).
Liquid Elastomeric Polymer	Fresh Water	Patent discloses several compositions, including polyurethane elastomers.

The asphaltic compositions considered in the preliminary studies for tailings impoundment liners included asphalt membranes, asphalt/fabric membranes, asphaltic concretes, and soil asphalts. These compositions are described below.

Asphalt Membranes

Rubberized Asphalt Membranes

Hot-sprayed membranes are prepared by heating a selected asphalt to 220-240°C and applying it to a prepared subgrade with a spray bar system. The selected asphalt must meet the temperature requirements for the application, be flexible at the lowest expected temperature, and be sufficiently viscous at the highest expected temperature to resist flowing from sloped sections (The Asphalt Institute 1976). Because rubber decreases the effect of temperature on asphalt viscosity (Dunning 1975), these temperature requirements can be met by adding small quantities (2-3 wt%) of rubber to the asphalt. The elastic properties of rubberized asphalt make this material better suited to conditions, such as subsidence and temperature cycling, expected at tailings repository sites.

Rubberized asphalts are also being developed to prevent crack propagation between layers of airport runways, and they have been used as water barriers beneath highway surfaces. The membranes are placed in two or more layers (lifts) to seal pinholes formed by escaping moisture in the first lift. Typically, the composition is applied at a rate of 7 L/m² to achieve a thickness of 5-8 mm. The membranes must be protected by a layer of soil before heavy equipment is operated above the liner surface (Stewart 1978; Middlebrooks et al. 1978).

Catalytic Airblown Asphalt Membranes

Catalytic airblown asphalt membranes use high-softening-point asphalts to form a membrane seal. Because this type of liner is designed for applications where it will be exposed to solar heating (e.g., canal linings or roofing applications), high-temperature creep properties are very important. The asphalt is heated and sprayed in place as above, but it requires

a catalyst to modify the viscosity properties of the asphalt for application.

Low-temperature ductility is also a concern with these asphalts. Haxo and White (1976b) exposed a catalytic asphalt liner to landfill leachate for 1 year and found that the membrane did not deteriorate visibly and absorbed 2.9% of the leachate. No firm permeability data were reported, but a significant increase in low shear-rate viscosity was noted. Pre-exposure permeability of catalytic membranes is less than 10^{-9} cm/s (Haxo 1976a). Because of the apparent potential of this material, catalytic airblown asphalt was selected for further study.

Asphalt Emulsion on Fabric

These membranes are created by placing fabric mats on the subgrade, sewing them together to form a single fabric liner, and applying an asphalt emulsion to seal the fabric. The asphalt is occasionally chip-sealed to provide additional protection from mechanical damage (The Asphalt Institute 1976; Clark et al. 1974). Permeabilities of less than 10^{-9} cm/s have been reported for this type of liner (Haxo and White 1976a).

This form of asphaltic membrane has several limitations that affect its suitability for long-term service, and the small amount of asphalt applied is a primary concern. Haxo et al. (1979), Haxo and White (1976a), and Haxo et al. (1977) report testing an 0.8-cm asphalt emulsion/fabric liner. If a hole develops in the asphalt and exposes the fabric to potentially corrosive materials or if the fabric is attacked by elements in the soil, an unsupported 0.8-cm asphalt liner would be required to withstand the tensile and shear stresses and hydrostatic pressure of the tailings impoundment. Oxidation, and resulting loss of flexibility, of the thin membrane is also a potential failure mechanism. The oxidized asphalt could delaminate from the fabric, resulting in the failure mechanisms outlined above. In addition, the asphalt emulsion can be applied only at temperatures above freezing. Because of these limitations, asphalt emulsion on fabric was excluded from further study.

Asphalt Concretes

Paving Concrete

Asphaltic compositions in this category are designed for roadway construction. Typically, paving concrete consists of 4-6 wt% asphalt; the remainder is carefully graded aggregate. Occasionally, rubber is added to decrease the effect of temperature on the asphalt properties. Because these concretes are designed for traffic use, they include 6-10 vol% voids so the concrete can move under pressure exerted by vehicular traffic without cracking. Based on the relatively high void content and the failure of this style of liner reported by Haxo and White (1976a) after exposure to landfill leachate for 1 year, this material received no further consideration.

Hydraulic Asphalt Concrete

Hydraulic concretes are similar to paving concretes; however, asphalt content is increased to 6-10% (Middlebrooks et al. 1978), voids are reduced to <4%, and aggregate contains smaller, more rounded rock and mineral fill. Permeabilities to 10^{-10} cm/s have been reported for these materials (Stewart 1978; Hickie 1976). To prevent seepage through the joints of adjacent lifts, hydraulic concretes are laid in two lifts and joints are staggered from 2 to 6 feet. These materials passed a 1-year exposure test to landfill leachate conducted by Haxo and White (1976a). Although the installed cost may be higher than for asphaltic membranes, hydraulic concrete appeared to warrant further testing.

Asphalt/Rubber Admixture

An asphalt/rubber admixture is a newly developed type of liner containing asphalt and graded rubber particles (from discarded tires) mixed in proportions of 75% asphalt and 25% rubber. The advantages of this material include the effects of rubber on asphalt discussed above; however, it differs from rubberized asphalt in rubber concentrations. The rubber acts as an elastomeric aggregate and allows the membrane to deform more than rock aggregate concretes. Volume changes during freeze/thaw of the liner due to

water absorbed by the rock aggregate are eliminated. Frobel et al. (1977) used this material at an application rate of 4.5 L/m^2 and reported satisfactory results. Permeability is 10^{-7} cm/s although experimental error is suspected and permeability may be lower. Because of the marked difference between the asphalt/rubber admixture and other liners selected for further testing, it was chosen for laboratory screening instead of the rubberized asphalt membrane.

Soil Asphalt

Soil asphalts consist of local sands and asphalt mixed in proportions of 94% sand and 6% asphalt. These materials are not acceptable for liner applications due to high permeability-- 10^{-3} cm/s (Haxo 1976a). This high permeability is caused by the similar sizes of the aggregate material, which complicates compaction and leaves a significant void content. Haxo and White (1976a) reported failure of a soil asphalt membrane exposed to landfill leachate for 1 year.

CONCRETES

Portland cement-based concretes have been used for many years to line various water impoundment facilities, but these admixtures exhibit several limitations with respect to mill tailings. For example, they are inherently susceptible to spall damage from exposure to freeze/thaw cycling, cracking due to volume changes experienced under repeated wetting and drying, and corrosion from the SO_4^{-2} ions present in several of the tailings leachates. They would require reinforcement if installed where significant substrate subsidence is expected. Water permeability of these concretes is on the order of 10^{-4} cm/s .

For tailings impoundments where SO_4^{-2} ions are not present, little ground motion is expected, annual precipitation is small, and subgrade water content is nearly constant, concretes that have been produced for use on bridge decks could be considered as liner materials. These concretes were developed to minimize damage from freeze/thaw cycling and to protect the steel reinforcement structure from corrosion by de-icing chemicals.

These specialized concretes are termed internally sealed concrete, polymer-impregnated concrete, polymer-portland cement concrete, and polymer concrete. However, because these materials have not been developed for large-scale use in ponds, and because of the strength and cracking limitations described below, none could be considered for laboratory testing.

Internally Sealed Concrete

Internally sealed concrete (ISC) is an admixture of portland cement, aggregate, and wax spheres. After the concrete is cast and partially cured, heat is applied to the top surface, melting the wax. Ideally, this seals the inherent interconnected pores in the paving. Typical specifications for these admixtures are presented in Table 4.

TABLE 4. Internally Sealed Concrete Specification

<u>Item</u>	<u>Specification</u>
Admixture Density	446 kg/m ³
Aggregate Content	60 Volume %
Wax Content	7.8 Volume %
Entrained Air	4-6 Volume %
Water-Cement Ratio	0.47
Wax Specifications	
Density--	60 kg/m ³
Sphere Size--	-20 + 80 Mesh
Wax--	75% Parrafin 25% Mortan

During curing, electrical heaters are placed on the surface of the concrete to melt the wax. To achieve the best results, the recommended minimum temperature in the concrete during the heat treatment is >85°C.

Among the concerns about this concrete sealing mechanism are stresses induced by the thermal gradients present in the concrete and by the thermal

expansion differences between the cement paste and the aggregate. Limestone aggregate is recommended to minimize the latter stresses; however, the acidic nature of the tailings leachate would dictate selection of an alternative material. These concretes demonstrate about 25% reduced compressive strength (Tyson 1978).

Polymer Impregnated Concrete

Polymer impregnated concrete (PIC) materials are designed to protect the portland cement from chemical attack, reduce freeze/thaw failure, and increase the mechanical properties of the concrete. As with ISC, this is accomplished by filling the pores--in this case with a polymer--as follows:

1. cast the concrete
2. dry the concrete to reduce water in the concrete pores
3. saturate the concrete with a monomer
4. polymerize the monomer.

Typically, liquid monomers are used but gaseous monomers are possible. All cements, aggregates, and admixes are compatible with this technique.

The strength and durability of this compound is clearly a function of the percentage of concrete voids filled with the polymer or, rephrased, a function of the amount of water removed from the cast concrete. Because water removal requires that the concrete be heated, the process is complicated for large applications.

Three methods of polymerizing the monomer are suggested: thermal-catalytic polymerization, promoted-catalytic polymerization, and exposure to gamma radiation from a ^{60}Co source. The first method requires that heat be applied to the saturated concrete, actuating the catalyst compound to polymerize the monomers. The promoted-catalytic method is similar but occurs at ambient temperature. Limitations of this concrete compound include:

- saturating the sloped concrete sections with the low-viscosity monomer solution

- inhomogeneities in the polymer coating
- stresses induced in polymers caused by the inherent volume reduction during polymerization
- cracks forming in the concreted after the treatment that would expose untreated concrete to the tailings leachate.

These compounds are reported to have permeabilities on the order of 10^{-10} cm/s (Hoff 1977).

Polymer-Portland Cement Concrete

Polymer-portland cement concrete (PPCC) compounds are designed to improve the same characteristics of portland concretes as PIC as well as concrete shear bond, bond-to-aggregate, tensile strength, and flexural strength. Unlike PIC, the polymeric materials are included in the cement/aggregate admixture prior to casting. Two types of PPCC were considered: those using premix and postmix polymers.

Premix PPCC is the most common form. The monomers are polymerized prior to addition to the concrete/aggregate admixture; and the polymers are generally added as latexes or polymer dispersions and result in distinct polymeric phases in the concrete formed. These compounds are moist-cured for one to three days and allowed to dry under ambient conditions.

Because the polymers are usually added as water-soluble compounds during mixing, prolonged contact with water leads to their dispersion. This eventually would result in a porous concrete with properties similar to ordinary portland cement concrete.

Limited success has been achieved with postmix PPCC. Problems include interference with portland cement hydration, chemical reactions between the cement and monomer, difficulty in producing a homogeneous monomer dispersion, poor bonds between the polymer and aggregate, and achievement of complete polymerization (Hoff et al. 1977).

Polymer Concrete

Polymer concrete (PC) was the most promising of the nonasphaltic concretes considered. This material is a polymer-aggregate admixture and uses the same mixing and installation equipment as portland cement concrete. PC exhibits much greater chemical stability, compressive and tensile strengths and freeze/thaw resistance than portland cement concretes and should have very low permeabilities (Hickey 1969; Hoff et al. 1977).

These concretes are made with a variety of resins or monomers and can be adapted to the required application. Aggregates should be dried to less than 2% moisture for best service. PC contains between 5 and 30 wt% monomer. The catalyst-promoter polymerization technique described in the PIC section is recommended (Hoff 1977).

PRESSURE GROUTING FOR LINER FORMATION

The possibility of creating an effective candidate liner for uranium-mill tailings leachate by injecting portland cement or epoxy-based grouts into the subgrade has been evaluated by Tamura and Boegley (1983) for the UMTRA Program and thus was not included in the PNL liner program.

GEOMEMBRANES

Polymeric materials in the form of geomembrane liners were also considered. These flexible lining materials can be classified generally as elastomers (rubbers) or plastics. Major advantages include low permeability and the ability to conform to changing substrate conditions caused by freeze/thaw or wet/dry cycling, sink-hole formation, subsidence, etc. Liners are delivered in large sheets, usually containing several factory seams, and are available in a variety of thicknesses and colors, with or without fiber reinforcement (scrim), and in special compositions modifying the basic polymer to meet specific exposure conditions (Stewart 1978).

These materials require particular care in relation to site selection, ground preparation, and installation. To maximize the potential effectiveness of the liner, potential liner-compromising mechanisms must be minimized. These liners can be damaged by animals or microbes; operation of heavy equipment; poor seaming techniques; stresses that exceed the tensile capabilities of the material during installation or subsequent ground subsidence; perforation by growing plants or sharp objects beneath the liner; potential buildup of gas pressure under the liner for various reasons; and excessive reverse hydrostatic pressure caused by a high water table (Williams 1978; Stewart 1978; E. I. du Pont A; E. I. du Pont C).

Proper design of the tailing impoundment can reduce these potential failure modes. A 15- to 30-cm layer of sandy soil or tailings placed over the liner will protect it from damage by equipment or animals and would be required for ozone or ultraviolet-sensitive polymers. Biocides or fungicides can be incorporated into the liner during initial processing to protect it from microbial attack, and no liner polymer that would nourish microbes should be selected. Seams constructed in the field are less reliable than those done by the supplier and should be kept to a minimum; however, the selected liner should demonstrate good field-seaming characteristics and each joint should be checked by conventional techniques.

The liner can be protected from overstressing by a number of techniques. The ground beneath the liner can be compacted to prevent sink-hole formation; installation crews can ensure that the liner is not stretched during installation; liners can be purchased with internal fabric reinforcement (scrim); and the slope of impoundment walls can be limited to 1 m vertical and 2 m horizontal. It is also recommended that any wall seams be oriented so that they run vertically (Schultz and Miklas 1980).

In addition to precompaction, several other preparations are necessary prior to liner installation. The ground must be sterilized to prevent vegetation from penetrating the liner, and all organic materials must be removed to prevent gas-bubble formation beneath the liner. In areas where high winds or widely fluctuating water tables are expected, vents and

sloped pond bottoms as well as scrims are recommended to prevent damage from gas accumulation (Lubina, Hovater and McCready 1979; E. I. du Pont C). Scrims are also recommended when high water tables can produce excessive hydrostatic pressures beneath the liner.

It is generally thought, because of limited historical use data in comparison with asphalts and clays, that polymeric geomembranes could not be considered a long-term (1000-year) barrier for uranium mill tailings. As a basis for comparison, however, one of the more promising materials discussed below was selected for laboratory testing.

Elastomers

Butyl Rubber

Butyl rubber is a polymer containing 97% isobutylene and 3% isoprene. This material has demonstrated lifetimes of greater than 20 years lining potable water reservoirs (Stewart 1978). Butyl rubber has good weather resistance, is serviceable between -7 and 120°C, will elongate greater than 300% and recover in this temperature range, has low water absorption and permeability [1.1×10^{-11} cm/s (Haxo 1976a)], and is available in 0.5- to 3.2-mm (20- to 125-mil) thicknesses with or without scrim (Stewart 1978; Williams 1978; Haxo 1976a).

Haxo et al. (1979) demonstrated that the properties of butyl changed little when the material was exposed to landfill leachate for 8 months (Table 5); and the results of preliminary tests with a HNO_3 solution (1.5 pH) are shown in Table 6 (Haxo et al. 1977; Haxo et al. 1979; Haxo 1976b). However, the material has low resistance to hydrocarbons; requires a difficult, two-part seaming procedure that is highly weather-dependent; and is less resistant to embrittlement by radiation damage than other elastomers considered. The pertinent characteristics of butyl rubber are presented in Table 7. Other elastomeric materials have improved characteristics when exposed to conditions expected in a uranium-mill tailings repository.

TABLE 5. Summary of the Effects of Immersion of Polymeric Membrane Liners in Leachate for Eight Months

Polymer	Number of Liners in Test	Absorption of Leachate, %	% Original Value for Unexposed Membrane			Change in Hardness, Points
			Tensile Strength	Elongation	S-200	
Butyl Rubber	1	1-2	90-97	104-106	85-86	0
Chlorinated Polyethylene	3	8-10	80-115	64-135	84-123	-5 to -1
Chlorosulfonated Polyethylene	3	13-19	82-124	97-107	47-113	-20 to -4
Elasticized Polyolefin	1	0.1	86-94	91-92	102-106	0
Ethylene Propylene Rubber	5	1-13.5	90-91	76-138	98-220	-1 to +2
Neoprene	4	1-19	69-100 ^(a)	82-103 ^(a)	79-102 ^(a)	-11 to +5
Polybutylene	1	0.1	96-99	96-97	99-103	-3
Polyester Elastomer	1	2.0	99-115	101-108	95-110	-4
Polyethylene	1	0.6	110-180	96-181	100-116	-7
Polyvinyl Chloride	7	1-3	91-110	98-129	76-102	-2 to +1
Polyvinyl Chloride plus Pitch	1	6.0	92	109-133	93-108 ^(b)	-2

(a) Data on fabric-reinforced neoprene liner number 42 were not included.

(b) S-100 - unexposed specimen broke at 150% elongation.

TABLE 6. Effect of Immersing Liner and Sealing Materials in Hazardous Wastes--A Preliminary

(Unit: % Increase in Area of Portion of Strip Immersed)

Item	ID Number	Strong Acid		Strong Base		Tank Bottom		Load Wastes		Sat/Unsat Oil ^(e)	Pesticide Weed Killer
		HFL ^(a)	HNO ₃ ^(b)	Stop Water	Spent Caustic	"Weed Oil" ^(c)	Tergol Clay	Low Pb Washing ^(d)	Gasoline Wash Water		
Polyvinyl Chloride (PVC)	10	CC	NVC	HDS	NVC	NVC	HDS, -17	NVC	NVC	HDS, -10	NVC
	11	NVC	NVC	HDS	NVC	NVC	HDS, -12	NVC	NVC	HDS, -5	NVC
	17	NVC	NVC	HDS	NVC	+11	HDS, -10	NVC	NVC	HDS, -6	NVC
	40	NVC	NVC	HDS	NVC	+24	SHD	NVC	NVC	SHD	NVC
	RN 49	---	---	---	---	+3	---	---	---	---	---
	59	NVC	NVC	HDS	NVC	+26	-6	NVC	NVC	SHD	NVC
	67	NVC	NVC	HDS	NVC	+5	SHD, -6	NVC	NVC	SHD	NVC
	RN 71	NVC	(g)	HDS	NVC	CC	HDS, -8	NVC	NVC	HDS, -6	NVC
Chlorosulfonated Polyethylene	RN 6 ^(f)	NVC	NVC	NVC	NVC	RVD	NVC	NVC	NVC	40	NVC
	RC 50	---	---	---	---	6	---	---	---	---	---
	55	NVC	NVC	NVC	NVC	---	NVC	NVC	NVC	36	---
Chlorinated Polyethylene (CPD)	12 ^(f)	NVC	NVC	NVC	NVC	RVD	6	NVC	NVC	11	NVC
	38	NVC	NVC	NVC	NVC	RVD 92	NVC	NVC	NVC	16	NVC
	RN 39	NVC	NVC	NVC	NVC	RVD	NVC	NVC	NVC	9	NVC
	RP 48	---	---	---	---	RVD ^(h)	---	---	---	---	NVC
	RP 73	NVC	NVC	NVC	NVC	---	NVC	BLS	NVC	CC, 2	---
Ethylene Propylene Rubber (EPDM)	8	NVC	NVC	NVC	NVC	75	24	NVC	NVC	77	---
	26 ^(f)	NVC	NVC	NVC	NVC	90	60	NVC	NVC	70	NVC
Polychloroprene (Neoprene)	9	CC	SCR	CC	NVC	111	77	NVC	NVC	42	---
	37	NVC	NVC	NVC	NVC	67	NVC	NVC	NVC	21	NVC
	RN 42	NVC	RVD	NVC	NVC	REV	NVC	NVC	NVC	5	NVC
	43	---	---	---	---	10	13	---	---	48	NVC
	RP 47	---	---	---	---	6	---	---	---	---	---
	RN 56	NVC	NVC	SHD	NVC	100	3	NVC	NVC	10	NVC
	RP 74	NVC	NVC	(h)	NVC	NVC	NVC	NVC	NVC	NVC	---

TABLE 6. Continued

(Unit: % Increase in Area of Portion of Strip Immersed)

Item	ID Number	Strong Acid		Strong Base		Tank Bottom		Load Wastes		Sat/Unsat Oil ^(e)	Pesticide Weed Killer
		HFL ^(a)	HNO ₃ ^(b)	Water	Caustic	"Weed Oil" ^(c)	Tergol Clay	Low Pb Washing ^(d)	Gasoline Wash Water		
Butyl Rubber	22	NVC	NVC	NVC	NVC	68	64	NVC	NVC	69	NVC
	24	NVC	NVC	NVC	NVC	91	59	NVC	NVC	89	---
	44	---	---	---	---	76	13	---	---	94	---
	RN 57 ^(f)	NVC	NVC	NVC	NVC	67	20	NVC	NVC	40	NVC
Elasticized Polyolefin	36	NVC	NVC	NVC	NVC	49	17	NVC	NVC	32	NVC
	41	NVC	NVC	NVC	NVC	42	18	NVC	NVC	24	NVC

(a) Hydrofluoric

(b) Hydrofluoric, acetic, and nitric acids.

(c) "Weed oil" (highly aromatic) 30%; water 70% (tank bottom waste).

(d) Low-lead gas washings.

(e) Oil pond 104.

(f) Materials incorporated in cells.

(g) 0.10 in. swell in each direction, curled, and delaminated.

(h) Complete loss of tear strength.

(i) Neoprene in satisfactory condition but reinforcing fabric (polyester) dissolved.

(j) Black rubs off, indicating possible dissolving of the liner.

ABBREVIATIONS:

ABS - Absorbed waste

BLS - Blistering of surface

CC - Color change

DEL - Delaminated

DIS - Dissolved or disintegrated

ABBREVIATIONS (cont'd):

HDS - Hardened and shrank

NVC - No visible change

RC - Reinforced and cured

REV - Reverted

RF - Reinforced with fabric

RN - Reinforced with nylon

RP - Reinforced with polyester

RVD - Removed specimen from waste because of excessive swelling or disintegration

SCR - Surface cracking and hardening

SFT-1 - Softened above waste

SFT-2 - Softened in waste

SHD - Slightly hardened

SHR - Shrank

SS - Slightly swollen

TABLE 7. Properties of Polymeric Liner Membranes Installed as Barriers

Cell Number Liner Number	1,19 21		2,20 17		3,21 7		4,22 6		5,23 16		6,24 12	
Material	Polyethylene		Polyvinyl Chloride		Butyl Rubber		Hypalon, with Nylon Scrim		Ethylene-Propylene- Diene (EPDM) Rubber		Chlorinated Polyethylene	
Thickness, mm (0.0001 in.)	0.25-0.30 (10-12)		0.51-0.53 (20-21)		1.55-1.65 (61-65)		0.81-0.91 (32-36)		1.24-1.35 (49-53)		0.79-0.81 (31-32)	
Coefficient of Water Per- meability, cm/s			7.3×10^{-13}		1.1×10^{-11}		3.6×10^{-12}		2.3×10^{-11}		2.0×10^{-12}	
Water Absorption, %												
2 h @ 100°C	-0.61		2.15		0.17		7.17		0.47		2.93	
7 d @ 25°C	0.38		0.95		0.18		2.04		0.61		1.43	
70 d @ 25°C					0.52		4.52		1.90		5.31	
Puncture Test, (a) 25 mm/min, maximum force, N (lb) elongation, mm (in.)					149 (33.5) 29 (1.14)		131 (29.5) 26 (1.01)		141 (31.6) 35 (1.38)		150 (33.8) 26 (1.03)	
Puncture Test, 500 mm/min, maximum force, N (lb) elongation, mm (in.)	61.9 (13.9) 19 (0.76)		115 (25.8) 18 (0.69)		199 (44.8) 31 (1.22)		146 (32.9) 15 (0.60)		175 (39.4) 37 (1.44)		209 (47.0) 26 (1.04)	
Splice Strength, peel, kN/m (lb/in.) shear, kN/m (lb/in.)	2.73 (15.6) 3.54 (20.2)		0.70 (4.0) 6.51 (37.2)		0.66 (3.8) 5.25 (30)		5.25 (30) 8.75 (50)		0.44 (2.5) 2.56 (14.6)		1.75 (10) 9.98 (57)	
Hardness, Shore A												
instantaneous	98		81		55		81		57		85	
10 s	98		76		51		79		54		87	
Grain Direction	With	Cross	With	Cross	With	Cross	With	Cross	With	Cross	With	Cross
Modulus @ 100%												
MPa	8.76		8.69		2.41		6.90		2.41		8.41	
(lb/in. ²)	(1270)		(1260)		(350)		(1000)		(350)		(1270)	
	7.10		7.79		2.02		5.93		2.41		3.59	
	(1030)		(1130)		(290)		(860)		(350)		(520)	

TABLE 7. Continued

Cell Number Liner Number	1,19 21		2,20 17		3,21 7		4,22 6		5,23 16		6,24 12	
Material	Polyethylene		Polyvinyl Chloride		Butyl Rubber		Hypalon, with Nylon Scrim		Ethylene-Propylene- Diene (EPDM) Rubber		Chlorinated Polyethylene	
Modulus @ 200%, MPa (lb/in. ²)	10.1 (1470)	7.24 (1050)	14.3 (2080)	12.8 (1850)	5.31 (770)	4.21 (610)	11.8 (1710)	9.17 (1330)	5.24 (760)	5.24 (760)	12.5 (1820)	5.79 (840)
Modulus @ 300%, MPa (lb/in. ²)	11.6 (1680)	7.72 (1120)			8.48 (1230)	6.90 (1000)			7.72 (1120)	7.72 (1120)	17.0 (2460)	8.27 (1200)
Tensile Strength, MPa (lb/in. ²)	11.7 (1700)	17.9 (2590)	18.2 (2640)	17.4 (2520)	9.93 (1440)	9.86 (1430)	13.2 (1920)	11.1 (1610)	10.4 (1510)	9.93 (1440)	17.0 (2460)	14.3 (2080)
Elongation, %	320	690	270	290	360	430	250	250	420	400	300	520
Set, %	177	667	68	77	15	18	115	106	13	19	199	230
Tear Resistance, (b) kN/m (lb/in.)	72.6 (416)	63.0 (360)	61.6 (352)	55.5 (317)	31.5 (180)	31.5 (180)	56.0 (320)	49.0 (280)	31.7 (181)	31.7 (181)	47.2 (270)	42.0 (240)
Creep Test (c) Load, N (lb)	7.78 (1.75)	7.78 (1.75)	14.0 (3.15)	12.7 (2.85)	14.0 (3.14)	12.5 (2.82)	25.3 (5.68)	29.8 (6.7)	8.18 (1.84)	6.89 (1.55)	2.22 (5.0)	14.1 (3.18)
• hours to failure	>72 NF	>72 NF	26	12	>94 NF	>94 NF	>96 NF	2.2	>143 NF	>143 NF	31	47
• elongation, %	13.3	47.6	194	138	108	111	194	253	112	102	186	344
• initial stretch, 1 min, %	5.7	7.7	73	51	84	83	15	17	79	78	62	129
• set, %	9.5	19			5.4	5.2		100+	5.6	4.7		176
• creep, cm/cm, 100 h	0.10	0.52	3.88	7.5	0.23	0.23	1.87	99	0.18	0.14	4.5	3.6

(a) Method 2065, Fed. Test Methods 101

(b) ASTM D624, Die C

(c) ASTM D674 - dumbbell specimens, restricted portion 51 mm (2 in.) long by 6.3 mm (0.25 in.) wide.

NF = no failure.

Ethylene Propylene Rubber (Nordel, EPDM)

Ethylene propylene rubber is a diene monomer similar to butyl rubber. Nordel is resistant to ozone and UV attack, is servicable between -50 and 100°C, and is not susceptible to damage from microbial attack. Nordel is available with or without scrim and in 20- to 60-mil thicknesses (E. I. du Pont A, B and C; Williams 1978). Although the durability of Nordel with hydrocarbons is superior to that of butyl rubber, it is not recommended for liner applications (E. I. du Pont A). As with all vulcanized materials, Nordel presents seaming difficulties; and Haxo et al. (1979) reported Nordel to be more affected than butyl rubber by landfill leachate (see Table 5). They also reported a significant loss of transverse flexibility during the exposure period. EPDM is slightly less resistant to radiation damage than hypalon^(a) (Mattia and Luh 1971), which is discussed in the following section. Properties of unexposed EPDM are listed in Table 4.

Chlorosulfonated Polyethylene

Chlorosulfonated polyethylene (CSPE), which is also called hypalon, is obtained by the reaction of polyethylene in solution with chlorine and sulfur dioxide. When delivered to the installation site, hypalon exhibits properties associated with plastics and self-vulcanizes upon exposure to UV radiation and soil moisture. The product has the following characteristics:

- is resistant to both ozone and ultraviolet light (UV)
- is very impermeable to water (3.6×10^{-12} cm/s for a 34-mil liner)
- performs satisfactorily from -18 to 120°C
- is readily seamed under wet conditions
- resists microbial attack

(a) Hypalon is a registered trademark of E. I. du Pont de Nemours and Company.

- is resistant to H_2SO_4 oxidizing chemicals (kerosene may be a problem, however)
- is available in 20,000-ft² rolls, 4 to 6 ft wide, with or without scrim (E. I. du Pont B; E. I. du Pont C; Haxo 1976a).

CSPE will burn as long as a heat source is supplied but will self-extinguish if the source is removed. Portions of the liner to be seamed must be shielded from UV radiation, and the material is relatively weak without fabric reinforcement (E. I. du Pont A; E. I. du Pont C; Stewart 1978). A variety of scrims is available, but polypropylene is recommended because its acid resistance is superior to nylon and polyester. Use of polypropylene scrim, however, requires an opaque hypalon because it is not UV-resistant (Edwald 1973).

Of all the elastomers studied, hypalon demonstrated the best resistance to hardening due to exposure to gamma radiation and remained relatively unchanged up to 10^7 R exposure (E. I. du Pont D). Table 8 presents the effects of gamma radiation on several characteristics of the polymers under consideration (Mattia and Luh 1971).

Haxo and White (1976a) determined that exposure to landfill leachate for 1 year causes a hypalon liner to swell and decreases the seam-peel resistance (see Table 9). However, the resulting seam-peel strength was on the order of the other polymers tested, and the hypalon seam-shear strength remained among the highest rated. As expected, the preliminary study (reported in Table 6) showed no damage to a hypalon liner exposed to a low pH solution (Haxo 1976b).

In addition, hypalon was chosen as part of the seepage barrier installed at the Cotter Uranium Mill for tailings impoundment. Lubina, Hovater and McCready (1979) reported that du Pont predicts that more than 40 years of service could be expected from an exposed hypalon liner, and indefinite life if the liner is protected by an earthen layer.

TABLE 8. Effect of Gamma Radiation^(a) on Physical Properties (Blodgett and Fisher 1969)

	PVC	High Density PE	Black XLPE	Unfilled XLPE	SBR	EPDM	EPM	Butyl	Silicone	Neoprene	Hypalon	Chlorinated PE
<u>Original Properties</u>												
200% Modulus, psi	2415	1000	1767	1260	580	1033	730	520	859	930	884	626
Tensile Strength, psi	2601	2213	2045	2272	1520	1443	872	798	1191	2544	2113	2170
Elongation at Break, %	250	640	270	480	460	470	300	450	290	550	560	670
<u>200% Modulus, % Retention After Irradiation, Rads</u>												
5×10^5	81	95	125	96	106	100	116	103	75	107	116	108
5×10^6	95	98	115	102	121	94	127	69	112	103	156	152
5×10^7	+	+	+	108	150	120	+	++	98	160	203	+
1×10^8	-	+	95	+	+	+	+	++	+++	-	-	-
<u>Tensile Strength, % Retention After Irradiation, Rads</u>												
5×10^5	80	96	122	102	98	104	101	96	76	104	106	112
5×10^6	88	98	112	97	100	97	106	58	100	98	113	98
5×10^7	61	123	101	70	82	93	119	++	100	77	124	135
1×10^8	-	118	95	59	40	79	90	++	+++	-	-	-
<u>Elongation, % Retention After Irradiation, Rads</u>												
5×10^5	100	103	104	90	93	111	96	93	107	96	89	99
5×10^6	80	103	96	96	96	102	81	87	90	93	86	63
5×10^7	40	-	48	58	70	47	41	++	34	46	59	18
1×10^8	-	2	37	25	33	32	26	++	+++	-	-	-

(a) Radiation Source - For exposures of 5×10^5 , 5×10^6 , and 5×10^7 , cobalt-60 source was used (gamma = 1.17 to 1.332 meV and beta = 0.31 meV, at a dose rate of 5×10^5 rads/h). For exposure of 1×10^8 , Esso Research and Engineering Company's radiation core in air and water was used, with the same dose rate as above.

+ Elongated less than 200%

++ Degraded (low strength due to chain scissions)

+++ Brittle

- No value reported

TABLE 9. Effect on Properties^(a) of Polymeric Membrane Liners of One Year of Exposure to Leachate from Simulated Sanitary Landfills (data in U.S. customary units)

Item	Exposure Time, Years	Polyethylene	Polyvinyl Chloride	Butyl	Chlorosulfonated Polyethylene	Ethylene Propylene Rubber	Chlorinated Polyethylene
Liner Number	---	21	17	7	6	16	12
Generator Number	---	19	20	21	22	23	24
Thickness, mils	0	11-12	20-21	61-65	32-36	49-53	31-32
	1	11	21	64	38	51	35
Tensile Strength, psi	0	2145	2580	1435	1765	1475	2270
	1	2465	2350	1395	1640	1455	1810
Elongation at Break, %	0	505	280	395	250	410	410
	1	560	330	410	300	435	400
Tensile Set, %	0	422	73	17	111	16	429
	1	432	57	14	106	12	208
S-200, ^(a) psi	0	1260	1965	690	1520	760	1330
	1	1205	1550	685	1245	740	1090
Tear Strength (Die C), ppi	0	390	335	180	300	181	155
	1	496	450	202	305	195	320
Hardness (Duro A - 10 s)	0	98	76	51	79	54	85
	1	---	64	50.5	64	51.5	65.5
Puncture Resistance ^(b) force, lb	0	11.9	25.8	44.8	32.9	39.4	47.0
	1	14.8	30.1	49.5	57.0	40.1	49.8
Elongation, in.	0	0.76	0.69	1.22	0.60	1.44	1.04
	1	0.80	0.70	1.20	0.88	1.18	0.98
Volatiles at 105°C	1	0.02	3.55	2.02	12.76	5.54	6.84
Seam Strength Peel, ppi	0	15.6	40	3.8	30.0	2.5	10.0
	1	10.3 ^(c)	5.1	2.9	3.4	2.0	5.1
Shear, ppi	0	20.2	37.2	30.0	50	14.6	57
	1	11.4	25.6	42.0	40.2	24.3	35

(a) Stress at 200% elongation.

(b) Rate of penetration of probe 20 in./min.

(c) Seam in polyethylene liner used in steel column. Tabs in liner specimens mounted in base were too short.

Because of its advantageous characteristics, chlorosulfonated polyethylene was selected as the geomembrane material for laboratory screening tests. The results of these tests were used as the basis for comparisons with asphalt and clay liners.

Neoprene

Neoprene is a chlorine-containing synthetic rubber designed to resist hydrocarbon solvents. Neoprenes are serviceable between -20 and 90°C, resist attack by ozone and microbes, and are available in 20- to 60-mil sheets (E. I. du Pont C). However, neoprene is susceptible to damage from acid and strong salt solutions expected in tailings impoundments and exposure to weathering when in contact with earth; and seaming must be performed above 10°C (Williams 1978; E. I. du Pont A). Radiation resistance is comparable to hypalon, but neoprene is still not suitable for this application (E. I. du Pont D).

Plastics

Elasticized Polyolefin (3110, 3111)

Elasticized polyolefin is a thermoplastic material with good aging characteristics and resistance to attack by oily solutions, microbes, and solutions between 2 and 13.5 pH. Because the material is not vulcanized, field heat-seaming is possible at rates to 360 m/hr. The material is extruded rather than calandered and is available in 6-m x 61-m x 20-mil sheets; however, because of the extrusion processing, it is not available with fiber reinforcement, which limits its applicability. Moreover, elasticized polyolefin must not be exposed to hydrocarbons, steep slopes (>33%), or impoundments where wet/marshy conditions exist. No radiation resistance data are available; and du Pont does not warranty this material (Williams 1978; E. I. du Pont B; E. I. du Pont C).

Polyethylene

Since polyethylene materials do not contain a plasticizer, extrusion-forming processes are required and sheet thickness is limited to 8 mm.

Polyethylene has poor resistance to UV exposure, requires an adhesive tape-style seal that is difficult to make, has limited resistance to punctures, and tends to crack in creases. Advantages such as chemical inertness and radiation resistance do not overcome the limitations of polyethylene (Stewart 1978; Williams 1978; E. I. du Pont D; Haxo 1976a); however, it is available in high- and low-density specifications.

High-density polyethylene appears to have excellent durability under conditions expected in uranium-mill tailings impoundments. Its lack of plasticizers eliminates concerns about microbial attack, and the material is resistant to degradation from hydrocarbons thought to be contained in tailings piles, high acidity, and UV radiation. Although these characteristics made high-density polyethylene worth considering for laboratory testing, it had limited use in the field at the time the selections were made, and chlorosulfonated polyethylene was chosen instead.

Polyvinyl Chloride

Polyvinyl chloride (PVC) is a calandered material that is available in a wide variety of thicknesses, colors, and compositions. PVC exhibits good resistance to puncture and microbes, has low permeability (7.3×10^{-13} cm/s for a 20-mil sheet), and is chemically stable in the environment expected in the tailings piles. Principal disadvantages include susceptibility to breakdown from exposure to UV radiation below 355 μm resulting in cross-bonding and increased stiffness, migration of plasticizers also resulting in increased stiffness, and moderate gamma exposures resulting in a compound that becomes soft and tacky. As with low-density polyethylene, the inherent limitations were serious enough to eliminate PVC from further consideration as a candidate liner material (Savartnick 1969; Stewart 1978; Williams 1978; E. I. du Pont D; Haxo 1976a).

Chlorinated Polyethylene

Chlorinated polyethylene (CPE) is a calandered thermoplastic available in 20- to 45-mil thicknesses with a polyolefin reinforcing sheet. The material is produced by chlorinating high-density polyethylene and can be seamed by adhesive solvent, solvent welding, or heat-sealing techniques.

CPE is not susceptible to ozone or UV radiation, resists hydrocarbons, is unaffected by microbes, and has a permeability of 2×10^{-12} cm/s for a 30-mil sheet. The material, however, is reported to have limited resistance to acids, requires reinforcement for use on slopes, and is not recommended on slopes of more than 2-m rise to 3-m horizontal. Susceptibility to acid attack precluded any further consideration.

LABORATORY LINER EVALUATIONS

At the completion of preliminary studies, eight of the more promising liner materials were selected for laboratory screening:

- hydraulic asphalt concrete
- catalytic airblown asphalt membrane
- asphalt/rubber membrane
- chlorosulfonated polyethylene (hypalon)
- natural soil from a typical disposal site
- soil amended with sodium bentonite
- soil amended with Saline Seal 100
- soil amended with GSR-60.

The clay liner materials listed above (sodium bentonite, Saline Seal 100, GSR 60) afforded a broad evaluation of sodium-bentonite-based liner substances. The eight candidate materials were placed in 0.6-m diameter exposure columns and subjected to conditions that accelerated the aging process. The performance of each material was evaluated by comparing changes in permeability with time of exposure. The two most suitable liners were identified for further laboratory evaluation and exposure to real-time conditions in the field.

ACCELERATED TESTING METHODOLOGY

Since EPA regulations (USEPA 1983) require that mill tailings disposal sites conform to 1000-year criteria, the approach chosen as the best way to determine long-term liner effectiveness was to accelerate the aging mechanisms while maintaining conditions as nearly representative of the field as possible. It was considered equally important to select destructive mechanisms that would be encountered in the field. For example, UV degradation of asphalt was not considered because the liner would be covered throughout its useful life. Likewise, freeze/thaw and wet/dry cycles were eliminated from practical consideration because a thick cover of tailings and soil over the liners would eliminate the possibility of such extremes.

The behavior of clay amendments and asphalt and geomembrane liners are such that different accelerated testing methodologies had to be employed. Asphalt and synthetic materials degrade primarily by oxidation, which causes them to become hard and brittle. The most serious degradation mechanism for clay materials is the ion-exchange reaction between Na present in the clays and Ca, which exists in high concentrations in uranium tailings leachates. This ion-exchange reaction decreases the swelling capabilities of the clay, increasing leachate permeability. Both asphalt and clay materials, however, are subject to the same physical stresses caused by ground subsidence when tailings are introduced to the disposal site. Gamma radiation effects over the 1000-year period were also considered.

Oxidation of Asphalts and Synthetics

Asphalt aging results in physical hardening and embrittlement of the liner materials; and the primary degradation known to occur due to aging is oxidation. When oxidized, asphalt is converted to longer-chain hydrocarbons through the formation of carbonyls. These longer-chained hydrocarbons are more viscous and brittle; thus, they are more prone to crack under physical stresses that might be experienced at a tailings site over a long period (i.e., 1000 years). It is important, therefore, to quantify the rate of aging reactions and predict the condition of the asphalt liner after the 1000-year period. For these reasons, a methodology was developed to accelerate the reaction rate over a reasonable period of time (3 months).

The objective of accelerated aging was to increase the degradation reaction rates of an asphalt liner under simulated mill tailings impoundment conditions. To accomplish this, certain exposure conditions were selected, based on past studies, to accelerate the reaction mechanisms. The reaction rate was determined and compared to those of less accelerated or normal conditions, and this comparison was used to quantify the expected performance lifetime.

Van Oort (1956) and Blokker and van Hoorn (1959) conducted studies on increasing the degradation rates of asphalt, and both used the following expression for the reaction rate:

$$R_A = k (C_{O_2})^n \quad (1)$$

where R_A = reaction rate of the component of interest

k = the temperature dependent reaction rate constant

C_{O_2} = concentration of oxygen

n = the dependency of oxygen on reaction rate (referred to as the order of reaction).

The reaction rate constant is dependent upon temperature according to Arrhenius law (Levenspiel 1972):

$$k = k_0 e^{-E_A/RT} \quad (2)$$

where k = reaction rate constant

k_0 = a proportionality constant

E_A = activation energy

R = gas law constant

T = temperature.

Since the reaction rate constant is temperature-dependent, Van Oort experimented with temperature to increase the reaction rate. By conducting aging experiments in the absence of light, Van Oort was able to determine the activation energy E_A for asphalt experimentally. By plotting $\ln k$ vs $1/T$, he derived the activation energy at 24 kcal/mole, which is a normal value for organic chemical reactions. At this activation energy, an increase in temperature of 10°C above ambient would be expected to increase the reaction rate by a factor of 4.3.

Blokker and van Hoorn (1959) expanded on Van Oort's investigation of asphalt aging in the dark. Whereas Van Oort concluded that the extent of

aging would be limited by diffusion to 7 μm , Blokker and van Hoorn demonstrated, and substantiated their demonstration with theoretical interpretations, that hardening can occur as deeply as 3 mm or more. Blokker and van Hoorn also increased oxygen pressure to accelerate the reaction rates. Depending upon the order of reaction in the reaction rate equation, the concentration of oxygen in the atmosphere can affect the reaction rate substantially.

Blokker and van Hoorn's experiments showed that n in the reaction rate equation was 0.6. This relationship indicates that the concentration of oxygen can be instrumental in affecting the aging rates of asphalt. Blokker and van Hoorn also found that the activation energy (the temperature dependence coefficient of the reaction rate constant) was 12 kcal/mole rather than 24 kcal/mole as measured by Van Oort. At this new activation energy, a 10°C rise over ambient will approximately double the reaction rate, which is sufficient to make temperature a good acceleration parameter. Based on Blokker and van Hoorn's data, temperature and oxygen partial pressure were selected as aging parameters for the PNL liner evaluation study.

To obtain the equivalent of 1000-year aging in 3 months, it was necessary to accelerate the reaction rate by a factor of 4000. Because temperature and oxygen alone were not expected to accelerate the aging reactions to the equivalent of 1000 years, another parameter was sought. Since most of the uranium mill tailings leachates have considerable oxidizing agents such as sulfates, it was considered possible that increasing the strength of these agents would increase the rate of chemical degradation.

Basic chemistry shows that the concentration of H^+ ions can increase the oxidizing strength of sulfate; therefore, an increase in the H^+ ion content in simulated leachate from a nominal pH of 2.5 to lower pH values had the potential of increasing the oxidizing strength of sulfates and other oxidizing agents. Thus, three accelerated aging parameters were identified for the uranium mill tailings liner evaluation: temperature, oxygen partial pressure, and acidity controlled by pH.

Ion Exchange in Clay Soils and Bentonites

The swelling property of a clay soil depends upon its ability to hold hydrated Na ions in its exchange complex; and aside from physical disruptions, the failure of a clay liner will result from loss of its swelling property. This can occur either through a cation-exchange reaction that replaces the Na in the exchange complex with divalent cations such as Ca, or as a result of the dehydrating effect of a high-salt solution that removes water of hydration from the exchangeable Na. Both effects are controlled by the concentration of salts in the contacting solution.

Concentration affects the exchange reaction because it is a mass-action phenomenon. In this testing program, it was necessary to establish conditions under which mass-action would occur over a short period of time and be representative of the results that might be obtained over the long term in an actual impoundment. To accelerate the ion-exchange reaction, a thin clay liner was used and pressurized air was introduced to the exposure columns to apply pressure to the leachate solution above the liner. In this manner, it was possible to measure the relationship between permeability and the number of pore volumes of tailings leachate that passed through the liner.

By equating permeability and number of pore volumes in Darcy's equation for saturated flow (Hillel 1980), exposure time in the laboratory can be related to equivalent time in the field by the relationship

$$\frac{t_1 (L_1/L_2)^2}{t_2 p_1/p_2} \quad (3)$$

where t = time

L = liner thickness

p = hydrostatic pressure

$_1$ = reference to laboratory measurements

$_2$ = reference to field measurements.

As the relationship shows, liner aging can be accelerated by exerting an increased hydrostatic pressure over a thin liner relative to that expected in the field.

Physical Stresses

When tailings are introduced to the disposal site, the soil mantle beneath the liner is expected to subside somewhat, and this subsidence will cause elongation or shifting of the liner along the sloped walls of the disposal site. Figure 6 is a model of a typical disposal site. Using this model, anticipated subsidence was calculated to be 33 cm. Assuming a 3:1 excavation slope with a tailings depth of 6 m, the maximum elongation a liner must withstand is 0.035 cm/cm, or 3.5%; therefore, this value was used in the accelerated tests for clay and asphalt and synthetic liners.

FIGURE 6. Typical Disposal Site for Predicting Subsidence

Radiation Exposure

Liner exposure to gamma radiation was estimated for a 1000-year period by making certain assumptions and forming relationships. Assuming the relaxation length of tailings is the same as concrete (6.5 cm at 0.35 MeV), a surface source was estimated for the tailings above the liner according to the relationship given by Foster and Wright (1973):

$$S = 1/2 S_v \lambda \quad (4)$$

where S = surface source, dis/min per cm^2
 S_v = volume source, dis/min per cm^3
 λ = relaxation length, cm.

The volume source was estimated from the highest gamma activities detected at the Salt Lake City, Shiprock, and Durango tailings piles for the ^{238}U decay chain. The volume source used in the exposure calculation was 36,000 dis/min per cm^3 (16 nCi/ cm^3), which is much higher than normally found in tailings. The absorbed energy from gamma radiation expected at this level incident to the liners over the 1000-year period is 10^4 R. A circular sample of each asphalt liner was irradiated from a point source of ^{60}Co at 10^5 R at the center, and no physical defects were observed on the liner materials.

EXPOSURE COLUMN DESIGN

Eight sealed columns were fabricated to produce a controlled environment in which the liners were exposed to accelerated aging conditions. The liner exposure columns, shown in Figure 7, were constructed of 61-cm (24-in.) 304L stainless steel pipe and were designed to simulate expected field conditions and to expose the candidate liners to the accelerated aging mechanisms described earlier. Design features are in Figure 8.

FIGURE 7. Accelerated Aging Laboratory Apparatus

FIGURE 8. Construction Details of Liner Exposure Columns

Impoundment characteristics that had to be simulated in the exposure columns included forces exerted by the tailings above the liner; the pressure head of tailings leachate in the impoundment, if any; and subsidence of the supporting substrate. The weight of the tailings above the liner was duplicated by applying a representative force on the simulated tailings in the column with the piston shown in Figure 8. The piston was fabricated from perforated plating so the simulated leachate could flow freely while the desired load was applied to the sand.

The pressure head of tailings leachate above the liner was simulated by applying gas pressure to the test cylinder. The effects of ground subsidence were simulated by a collapsible, water-filled bladder installed in the supporting sand beneath the liner. The bladder was constructed from two hypalon sheets clamped at the edges with polyvinyl-chloride rings. The space between the sheets was filled with sufficient water to produce the displaced volume required to model subsidence.

The bottom of each exposure column was sloped so fluid passing through the liner was directed to the drain. The welded bottoms of the columns, constructed of carbon steel, were coated with an asphalt layer or an epoxy resin for corrosion protection.

The exposure columns containing the asphalt and chlorosulfonated polyethylene liners were heated by circulating hot water through external copper coils, which were centered around the liner to provide uniform heating. The circulating water temperature was controlled by a heat pump or immersion heater system.

The exposure columns were instrumented with moisture blocks above and below the liners to monitor the degree of saturation of the upper and lower column halves. In addition, the temperature of the asphalt and chlorosulfonated polyethylene cylinders was monitored by thermocouples placed in the simulated tailings above the liner. This was necessary because liner temperature is one of the most important variables controlling the oxidation rate and these data were required to predict liner aging.

SCREENING TEST RESULTS

The liners and supporting soil were sandwiched in the exposure columns between simulated tailings and a sand layer. The tailings above the liner in each of the columns were simulated with Wedron washed silica sand, Size No. 70, which was selected because it resembled the particle size distribution of samples removed from the New Rifle, Colorado, tailings pile (see Figure 2, Hartley et al. 1980) and it would not affect the chemical reactions between the leachate and the liner. The same sand was used beneath the liners to minimize absorption of leachate that passed through them.

Just below the asphalt and chlorosulfonated polyethylene liners, a 0.5-cm layer of soil was installed to subject the bottom surface of the liner to realistic conditions. Also, in every case the centerlines of the subsidence bladder and moisture blocks were placed 9.5 cm and 3.2 cm below the liners, respectively. In each of the cylinders, the sand beneath the liner was compacted with a vibrating rod.

The permeability values, k , calculated for the testing cycles are presented in Figures 9 and 10. These values were calculated using the Darcy equation for saturated flow:

$$k = \frac{\Delta V L}{A P t} \quad (5)$$

where k = permeability, cm/s

V = change in fluid volume, cm^3

L = liner thickness, cm

A = surface area of liner, cm^2

P = pressure difference across the liner, cm H_2O

t = permeability cycle duration, s.

Depending on the permeability of the liner, ΔV was measured by the volume of fluid collected below the liner during a permeability measurement cycle or by the loss in volume above the liner as measured by the manometer. Pertinent data for each permeability test (ΔV , P , t , etc.) are listed in Appendix A.

FIGURE 9. Permeability Performance of Asphalt and Hypalon Liners During Laboratory Testing Period

FIGURE 10. Permeability Performance of Clay-Amended and Natural Soil During Laboratory Testing Period

The change of permeability with exposure time is a good indication of the relative stability of the liners; however, when evaluating the effectiveness of liner materials of different thicknesses, it is not sufficient to compare permeability alone. In the field, a liner such as a soil amended with bentonite 10 to 20 cm thick need not be as impermeable as a thin membrane to be an effective leachate barrier. Therefore, the liners were also compared by averaging the permeabilities from the final 30 days and dividing the liner field thickness. The K/L factors, called the permeability effectiveness factors, for all liners are listed in Table 10.

TABLE 10. Anticipated Field Liner Permeabilities

<u>Liner</u>	<u>Average Final Permeabilities, K, cm/s</u>	<u>Assumed Field Thickness, L, cm</u>	<u>Effectiveness₋₁ Factor, K/L, S⁻¹</u>
Asphalt Concrete	$7(10^{-8})$	10	$7(10^{-9})$
Hypalon	$2(10^{-10})$	0.12	$2(10^{-9})$
Asphalt Rubber Membrane	$4(10^{-6})$	0.8	$5(10^{-6})$
Catalytic Airblown Membrane	$7(10^{-9})$	0.9	$8(10^{-9})$
Sodium Bentonite	$1(10^{-7})$	10	$1(10^{-8})$
Saline Seal 100	$8(10^{-6})$	10	$8(10^{-7})$
GSR-60	$6(10^{-6})$	10	$6(10^{-7})$
Soil (as a liner)	$1(10^{-5})$	10	$1(10^{-6})$

Asphalt and Synthetic Liners

With the exception of the CAA membrane, the permeability of each of the asphalt and synthetic liners increased during the laboratory testing period. A discussion of the performance of each of these liners follows; and the pressure, pH, and temperature of the exposure columns during the testing period are plotted in Figures 11, 12, and 13, respectively.

FIGURE 11. Oxygen Pressure to Which Asphalt and Synthetic Liners Were Exposed During Testing

FIGURE 12. Acidity Concentration During Testing of Asphalt and Synthetic Liners (includes pH values measured after test completion, June 30)

FIGURE 13. Exposure Temperature of Asphalt and Synthetic Liners During Testing

Asphalt-Rubber Liner

The asphalt-rubber composition evaluated in this study is currently being installed in the cooling-water lagoons at the Palo Verde Nuclear Power Station in Arizona. The mixture contained approximately 79% ASTM Type 1 Roofing Asphalt, 19% U.S. Rubber Reclaiming GP274 reclaimed tire rubber, and 2% high-boiling-point asphalt extender oil. The liner was fabricated on a specially treated paper backing at the University of New Mexico's Engineering Research Institute (UNMERI) and shipped to PNL. The asphalt application rate for this liner was 5.4 L/m^2 , the minimum application recommended by the UNMERI staff.

Visual examination of the liner after laboratory testing revealed tears at the outer edges of the exposure column. No evidence of failure was detected at the repaired tear in the liner center (the liner was torn and repaired during installation). Tears along the outer edges indicated

insufficient elasticity and strength when the subsidence bladder was collapsed near the end of the test. This event also produced the permeability increases shown in Figure 9. Another permeability increase was observed subsequent to acid addition to the tailings leachate and may indicate that the elastomer used in this liner is not compatible with highly acidic environments.

Hydraulic Asphalt Concrete

The asphalt concrete liner was produced by installing two 4.5-cm thick lifts of admixed asphalt above a cationic asphalt emulsion tack coat. Asphalt emulsion was also applied to the cylinder walls to bond the liner to the exposure cylinder. An AR4000 asphalt cement, containing 10% asphalt and 90% aggregate, was used to fabricate this liner. Specifications are provided in Appendix B.

The asphalt and aggregate were heated to 200°C and mixed in two 12.5-kg batches. After mixing, the admixture was reheated to 200°C and installed in the exposure column. Individual lifts were poured into the cylinder, leveled, and partially compacted by hand; then they were compressed to 78 psig in a hydraulic press. This pressure was used because it represents the compactive force developed in a 90-blow Marshall compaction procedure and because it represents the compressive action of a 5-ton vibratory roller. The lower asphalt concrete lift was allowed to cool for 30 minutes before the upper lift was installed.

The permeability of this liner increased early in the laboratory tests but stabilized at 10^{-7} cm/s. This result suggests that one (or more) of the liner components--either the asphalt cement, the asphalt emulsion used to seal the edges, or the aggregate--was degraded by the accelerated aging testing; however, comparison of the effectiveness of the liner permeability with that of other liners shows this material to be acceptable.

As expected, this asphalt concrete liner did not demonstrate a permeability increase in the subsidence test. No significant effect was expected because of the inherent strength of the 7.6-cm thick liner.

Catalytic Airblown Asphalt Membrane

The catalytic airblown asphalt (CAA) membrane, a hot-sprayed liner, was also applied in two lifts. Asphalt was placed in a heated reservoir and pressurized with nitrogen, which forced the CAA through a manually operated spray gun. The first lift was sprayed directly on the soil layer; the second lift was sprayed from an opposing direction to assure complete coverage. The application rate for the completed liner was 6.8 L/m^2 , and the CAA was sprayed at a temperature of 205°C . The specifications of this liner are listed in Appendix B.

Permeability measurements generated during tests of this liner displayed the greatest amount of data scatter. This variability is attributed to two factors. First, excess tailings leachate that collected in the bottom of the cylinders between permeability measurement cycles was included in the total cylinder volume change. Second, the liner was used as a gasket to seal the flanged connection at the center of the exposure column. Occasionally, leaks developed at this connection as the asphalt flowed to relieve stress applied by the bolts holding the flanges together. Gas pressure in the column pushed the simulated tailings leachate above the liner through these leaks. The low and high permeabilities measured before and after the subsidence test were caused by these mechanisms and are of questionable value.

Despite the erroneous readings on a few data points, enough permeability data were collected to demonstrate the stability of the CAA membrane when exposed to accelerated conditions. This was the only material in the asphalt and synthetic liner category that exhibited a permeability at the end of the exposure period that was not greater than the initial measurements. In addition, the permeability effectiveness factor ($K/L = 8 \times 10^{-9}$) was comparable to the best of the liner materials tested. No visible degradation was observed.

Chlorosulfonated Polyethylene

To represent geomembrane liners, a sheet of 1.5-mm chlorosulfonated polyethylene (hypalon) was placed above the 0.5-cm soil layer and sealed to the cylinder with silicone rubber cement. This liner consistently produced the lowest permeability measured; however, because of its thickness, the permeability effectiveness factor, K/L , of this membrane was within the same order of magnitude as the CAA membrane and the hydraulic asphalt concrete liner.

The subsidence test produced no permeability change and no visible degradation was observed, but salt deposits were noted at the outside edge of the liner. (This liner was also used as the center gasket; therefore, the outer edge was outside of the exposure column.) The salt deposits were attributed to leachate wicking along the fiber reinforcement. A slight increase in permeability with time was noted, indicating some decrease in stability.

Clay Liners

The four clay liners did not achieve the same initial permeabilities although each was prepared with the same 1:10 weight ratio of clay to soil. The differences in permeabilities were caused by several factors. A procedural error occurred during installation when the Saline Seal 100 and the GSR 60 liners were not moistened before compaction. In addition, the desired initial compaction could not be achieved because of fluid motion in the subsidence bladder. Finally, there were differences in particle size between the natural sodium bentonite and the polymer-treated bentonites. The latter were more granular and inhibited the passage of the polymer-treated bentonites into smaller pores of the soil. A thicker liner installed under field conditions should eliminate these problems. The pressure and pH histories of these liners are plotted in Figures 14 and 15.

FIGURE 14. Pressure History During Testing of Clay Liners

FIGURE 15. Acidity Concentration During Testing of Clay Liners (includes pH values measured after test completion, June 30)

Sodium Bentonite Liner

Natural sodium bentonite clay was mixed at a weight ratio of 1:10 with a soil sample from a prospective disposal site for the Durango Tailings Pile (Bodo Canyon, Site D). The soil-clay mixture was premixed, moistened, installed in a 2.5-cm layer, and manually compacted to the maximum attainable density. (The compaction process was hindered by the motion of the fluid in the subsidence bladder.) The liner-cylinder wall joint was sealed with a layer of pure sodium bentonite clay.

The permeability of the natural sodium bentonite liner increased initially, then decreased to roughly 10^{-7} cm/s. The increase was partially due to the early experimental variability described above, but may also indicate Na-Ca ion exchange early in the laboratory test. Permeability of this liner did not increase during the subsidence test; and this result supports the observation that this clay material was better distributed in the soil because of its small particle size and, therefore, produced a more stable liner. This liner was the most effective of the clay materials evaluated.

Saline Seal 100 and GSR-60 Liners

Saline Seal 100 and GSR-60 are bentonite materials supersaturated with sodium and treated with polymer to slow the $\text{Na}^+/\text{Ca}^{+2}$ ion-exchange reactions with leachate. The polymer is not expected to interfere with the coprecipitation of hazardous ions in the leachate.

The installation procedure for the processed bentonite liner was similar to that for the natural sodium bentonite liner. The same clay/soil ratio and edge sealing procedure were used, but these liners were not premoistened before compaction because of a procedural error. Native soils from the Bodo E and Long Hollow East Durango sites were used for the Saline Seal 100 and GSR-60 liners, respectively.

The permeability of the Saline Seal 100 liner decreased from its original, relatively high value and reached a plateau of 6×10^{-6} cm/s.

It remained at this level until the subsidence test, when the permeability increased as the liner was disrupted. A slow decrease in permeability after subsidence indicated that this liner exhibits some self-healing characteristics.

The permeability of the GSR-60 liner was nearly constant 3×10^{-6} cm/s from the beginning of the laboratory tests until the simulated subsidence. A large permeability increase was observed during the subsidence test because this membrane was also disrupted; and again, the clay demonstrated both a tendency to seal the liner following a disruptive incident and to reduce permeability following the subsidence test.

Natural Soil Liner

A control liner of soil from the Bodo E Durango disposal site was installed in one of the exposure columns. This liner was dry-compacted to a 2.5-cm thickness and sealed to the column with the sodium bentonite liner installation technique. The earth was passed through a 0.6-cm sieve to remove large rocks, which could have damaged a thin liner. No subsidence bladder was installed in this cylinder.

The natural soil liner was installed as a control to monitor any permeability change resulting from degradation. This liner displayed a generally increasing permeability trend throughout the tests.

Liner Costs

Installation costs were also compared. Charges were estimated by contacting vendors and installation contractors, who were asked to provide pricing information based on a $210,000\text{-m}^2$ impoundment at Clive, Utah. Estimates included material, transportation, and installation costs for each liner; however, variations in subsurface preparation requirements for these liners were not included. Cost estimates are presented in Table 11.

TABLE 11. Costs of the Candidate Liner Materials

Liner	Installation Rate	Installed Costs
Asphalt concrete	Two 5-cm lifts with cationic asphalt emulsion tack coat (3 L/m ²)	\$9.20/m ²
Chlorosulfonated polyethylene	0.76 mm (30 mil)	\$5.90/m ²
Asphalt-rubber	5.4 L/m ²	\$2.40/m ²
Catalytic airblown asphalt	9 L/m ²	\$2.60/m ²
Saline Seal 100	20 kg/m ²	\$4.70/m ² (a)
GSR-60	20 kg/m ²	\$2.70/m ² (a)
Sodium bentonite	20 kg/m ²	\$2.30/m ² (a)

(a) Assumes that 10₃ 2-cm (4-in.) layer of clay and soil is installed with soil at \$5.20/m².

Conclusions

The screening tests produced the following findings:

- Expected gamma irradiation in the field will not impair liner performance over the 1000-year period.
- Simulated subsidence indicated that asphalt concrete, chlorosulfonated polyethylene, sodium bentonite-amended soil, and probably the catalytic airblown membrane will yield the best performance.
- Hydraulic asphalt concrete, catalytic airblown asphalt, chlorosulfonated polyethylene, and sodium bentonite-amended soil are the liners with the best potential for field performance (as determined by permeability effectiveness factors, K/L).
- Catalytic airblown asphalt and soil amended with natural sodium bentonite are the most stable of the liners tested (as determined by the change of permeability with time).

- Initial permeability increases measured on the soil and natural sodium bentonite liners may indicate that rapid Na-Ca ion exchange is occurring. Since the overall performance was not affected, however, it may be assumed that the resulting shrinkage occurs vertically. This was confirmed by determining the extent of the ion-exchange reaction in subsequent studies.
- Asphalt-rubber and catalytic airblown asphalt are the least expensive of the asphalt and synthetic materials to install.
- Natural sodium bentonite and GSR-60 are the least expensive of the clay materials to install.

Based on the laboratory test results, expected field effectiveness and cost analysis, catalytic airblown asphalt and sodium bentonite-amended soil were selected for the fiscal year 1981 field studies and subsequent laboratory studies.

CONTROLLED AGING TESTS OF THE ASPHALT MEMBRANE

With the selection of the catalytic asphalt membrane as the most prospective liner in its group, further studies were required to quantify its expected performance lifetime. It was necessary to determine the penetration of reaction into the liner as well as the activation energy and dependency of pH and oxygen concentration on reaction rate; therefore, a series of exposure conditions were imposed.

Exposure Conditions

To select three values for each of the three accelerated aging parameters, a 3 x 3 x 3 testing matrix was used, resulting in 27 liner exposure tests. Since only eight exposure columns were available, however, a statistical method was used to select only key parameter combinations in the 3 x 3 x 3 testing matrix. This method, known as the fractional factorial matrix, requires measurements at 11 points rather than 27 (see Figure 16). With this statistical method, the high and low values of each

FIGURE 16. Fractional Factorial Matrix

parameter are selected for testing; and the liner is subjected to three exposure tests at the intermediate values of all three parameters to provide a statistical variation of the data. With eight exposure columns available, the 11 measurements could be made in two series of tests, leaving spares to rerun any cases that might be necessary.

The glycol cooling/heating coils on the exterior of the exposure columns were used to control temperature for the various points on the fractional factorial matrix. Intermediate and high temperatures (35 and 50°C) were maintained by electric heaters immersed in glycol that was continually recirculated through the heat transfer coils of the respective columns. The intermediate- and high-temperature cases were selected on the basis of anticipated reaction rate constants and the desire to avoid chemical reactions in the asphalt above 50°C that might not occur normally. The 20°C ambient temperature, which was maintained in the column by the heat pump, represented the equivalent ground temperature that might be expected below a uranium mill tailings pile.

The range of oxygen partial pressures for accelerated aging was based on ambient conditions, the maximum pressure rating of the exposure column, and an arithmetic average between the two. Therefore, 0.21, 1, and 1.7 atm were selected as the exposed oxygen partial pressures with a total pressure of 1.7 atm in all cases. The prescribed partial pressure of oxygen was maintained in the exposure column by two flow control valves from oxygen and nitrogen supply tanks. The intermediate and ambient oxygen partial pressures were more difficult to maintain because the flow of oxygen and nitrogen had to be balanced to match the leak rate of the exposure columns.

The asphalt liner was continually exposed to acidic leachate of the composition listed in Table 12. Acidity levels were maintained by periodic leachate sampling directly above the liner. As the pH slowly increased throughout the exposure period, additional sulfuric acid was introduced into the exposure column. The pH tended to increase because the leachate interacted with the exposure column, the simulated tailings, and possibly the asphalt liner itself.

TABLE 12. Simulated Tailings Leachate Composition

<u>Chemical</u>	<u>Concentration, g/L</u>
CaSO ₄	500.0
CaCO ₃	18.1
MgSO ₄	8.6
Na ₂ SO ₄	7.4
NaCl	7.4
Fe ₂ O ₃	2.4
NaCO ₃	2.3
Al ₂ O ₃	1.2

This situation posed a difficult control problem and required that various conditions be rerun in the second series of tests. The base case of pH = 2.5 was selected as the lowest measured pH from a variety of batch leaching samples of uranium mill tailings from Shiprock, Durango, and Salt Lake City. The pH values selected for the accelerated conditions were 2.0 and 1.5.

The column leaching studies indicated that the initial pore volume of leachate passing through Shiprock tailings had a pH of 1.8 and increased to higher pH values with subsequent pore volumes of leachate (Relyea and Martin 1982). The concern over maintaining accurate pH levels proved to be inconsequential, however, since it was discovered during asphalt liner analysis that acidity levels have an unmeasurable effect on the extent of aging.

Appendix C contains the pH curves measured for all asphalt samples during both series of tests. Temperature and oxygen exposure conditions are also given.

Permeability

Permeability was used to measure the immediate effectiveness of the asphalt liner. Because of the low permeability of the asphalt liner (10^{-9} - 10^{-10} cm/s), measurable volumes of leachate were collected infrequently; therefore, permeability was determined by changes in manometer levels during the exposure period. As indicated by the manometer, the time required for a permeability measurement was approximately 2 weeks.

Due to the chemical durability of the asphalt, accelerated aging conditions produced no measurable effect on permeability with time, even after the subsidence bladder was collapsed. Figure 17 represents the permeability measurements for three liners at normal, intermediate, and highly accelerated conditions. The flatness of the curves indicates that permeability cannot be used to assess the extent of aging in the asphalt liner and that additional analytical techniques are necessary to make such a determination. The permeability curves do, however, indicate that the

FIGURE 17. Permeability of Asphalt Liner at Normal, Intermediate, and Highly Accelerated Conditions

performance of the asphalt liner was not affected even for under maximum accelerated aging conditions. Permeability data for each run are given in Appendix C.

Analytical Results

Since premeability was unsatisfactory as a measure of the extent of aging, the formation of reaction products was analyzed as a function of depth into the liner. This information aided in determining whether aging was a near-surface phenomenon or whether it extended into the depth of the asphalt.

The asphalt membrane was sampled in 4-m increments with a microtome, which is generally used to obtain thin biomedical samples for analysis of animal tissue. To adapt microtoming for the asphalt study, special casting techniques were used to encase the asphalt sample in Paraplast^(a) (paraffin wax) or O.C.T.^(b)

Infrared spectrometry was selected to determine the extent of aging. This technique indicated how much of the asphalt had been converted to carbonyl forms, which are products of aging. The formation of carbonyls is important in the asphalt process because it occurs at the end of a hydrocarbon chain. These polar, oxygen-containing groups sometimes combine with other hydrocarbon chains, increasing the higher molecular weight fraction in the asphalt and the viscosity and brittle nature of the membrane. Experience with infrared spectrometry showed that all asphalt samples at various depths could be analyzed cost-effectively.

Separate infrared analyses of Paraplast and asphalt indicated that the absorption bands for these two materials are very similar; however, their similarities were not discovered until after the first series of tests had been cast and microtomed for analysis. For this reason, the second series of asphalt samples was cast in O.C.T., a material with absorption bands quite different from asphalt. Figure 18 shows the absorption bands and

(a) Paraplast is a registered trademark of the Lancer Division, Snerwood Medical, St. Louis, Missouri.

(b) O.C.T. (Optimal Cutting Temperature) is a registered trademark of the Labtek Division, Myles Laboratory, Naperville, Illinois.

FIGURE 18. Absorption Bands of O.C.T., Paraplast, and Asphalt

associated contributors for each of these materials. The carbonyl absorption band falls within the 1667 to 1800 wavenumber range, and the peak at 1600 wavenumbers is associated with C=C bonds from which carbonyls form.

Reaction rates for each sample were determined in the following manner. First, the contributions of the Paraplast or O.C.T. to the absorption

spectra were subtracted from the absorption band to obtain the true curve for the asphalt sample. Then, the aging reaction rates were determined by comparing the integrated values of these asphalt absorption curves related to various exposure conditions. The integrated absorption curves for each exposure condition were listed as a function of penetration into the liner as shown in Appendix C. The reaction rate at each depth was determined by rate of formation of carbonyls during the exposure period. The process can be expressed by the equation:

$$R = \frac{C_{C=O}|_f - C_{C=O}|_i}{t} \quad (6)$$

where R = reaction rate

$C_{C=O}|_f$ = concentration of carbonyls after exposure, indicated by the integrated absorption curve between 1667 and 1800 wavenumbers

$C_{C=O}|_i$ = initial concentration of carbonyls based on a derived relationship with the integrated absorption curve for $C=C$

t = exposure time.

Because the influence of the initial concentration of carbonyls was unexpected, the concentration of carbonyls ($C_{C=O}|_i$) was not measured before exposure. However, a mathematical relationship was derived to correlate the concentrations of $C=C$, which were measured with $C=O$. The expression was based on empirical data from the three tests in the center of the fractional matrix with identical exposure conditions. Then the reaction rates from Equation (6) were normalized to their respective, initial concentrations of carbonyls and plotted for the various exposure conditions as shown in Figures 19, 20, 21, and 22. Examination of these plots revealed some significant findings:

- temperature and oxygen are effective acceleration parameters
- acidity levels have no measurable effect on aging reactions
- no measurable reaction rate is observed under normal, nonaccelerated conditions

FIGURE 19. Reaction Rates at Normal and Highly Acidic Conditions. Liner designations correspond to data compiled in Appendix C.

FIGURE 20. Effect of Temperature on Reaction Rates at High Oxygen Concentrations. Liner designations correspond to data compiled in Appendix C.

FIGURE 21. Effect of Oxygen on Reaction Rates. Liner designations correspond to data compiled in Appendix C.

FIGURE 22. Reaction Rates at Intermediate Exposure Conditions. Liner designations correspond to data compiled in Appendix C.

- aging is extremely slow for asphalt, even under accelerated conditions
- aging reactions under accelerated conditions penetrate no deeper than 10 to 40 μm .

The basis for these conclusions is discussed below. The solid horizontal lines shown in Figures 19 through 22 are arithmetic mean values of the reaction rates measured at the near surface (10-40 μm). The standard deviation of the reaction rates beyond 40 μm were determined separately to define the bounds of the data scatter. The reaction rate near the surface was considered measurable if the mean value near the surface fell near or outside the standard deviation at greater depths.

Figure 19 illustrates two cases conducted at normal exposure conditions and one at high acidity and low temperature and oxygen conditions. No measurable reaction rates are discernible as a function of depth; therefore, we can conclude that aging reactions are not measurable under normal conditions during the 3-month exposure period. In addition, increased acidity in the leachate did not accelerate the aging reactions. Apparently the oxidizing nature of sulfuric acid is not a significant degradation mechanism for asphalts.

Figure 20 shows the effect of temperature on reaction rates for the same oxygen concentrations. The ratio of reaction rates for these two cases was $0.099/0.058 = 1.7$. A comparison of this low-temperature, high-oxygen reaction rate with the high-temperature, high-oxygen rate shown in Figure 21 results in an increased reaction rate near the surface by a factor of 0.31 (high-temperature, high-oxygen, Figure 21)/ 0.058 (low-temperature, high-oxygen, Figure 20) = 5.3 . Using the higher ratio of reaction rates, the activation energy in Equation (4) can be calculated to be 15 kcal/g-mole , which means that for a temperature increase of 10°C , the reaction rate increases by a factor of 2.3 . This is consistent with Blok-ker and van Hoorn's findings in their accelerated aging tests with pavement asphalts.

Figure 21 also shows the effect of oxygen concentration under constant temperature conditions. This comparison indicates that oxygen evidently has a significant effect on aging. From this plot, the value of n in the reaction rate Equation (3) is calculated to be 0.8. Blokker and van Hoorn determined n to be 0.6, so this finding was substantiated.

The penetration of aging reactions was limited to a depth of 10-40 μm in the surface of the 7000- μm thick liner. Blokker and van Hoorn reported that penetration of oxygen as deep as 3000 μm might be expected for thick samples. The apparent discrepancy may be due to the fact that the asphalt liner aging reactions were obtained under saturated leachate conditions, whereas Blokker and van Hoorn's studies were conducted in air.

Figure 22 shows the reaction rates at intermediate exposure conditions. These plots substantiate the quantitative effect of temperature and oxygen on reaction rate. The depth of penetration, 10-40 μm , was also substantiated by these measurements. Even though the data scatter was significant, the mean values agreed closely for all three exposed liners. The intermediate exposure results provide an important validation of the accelerated aging methodology and analysis.

Performance Lifetime

The calculated activation energy (E_A) and dependence of oxygen concentration on reaction rate (n) in the reaction rate equation determined the equivalent exposure period for the maximally exposed liner sample. With a temperature 20°C above ambient and 1.7 atm oxygen partial pressure, the reaction rate was increased by a factor of 30. In other words, the equivalent aging period of the liner over 3 months at these conditions was 7 years. Because acidity did not act as an accelerating aging parameter and only 40°C was achieved at the accelerated temperature (see Appendix C), the equivalent exposure period fell short of the desired objective. However, the expected performance lifetime can be extrapolated with some accuracy because the penetration of aging reactions was minimal.

At maximum exposure conditions, penetration of the reaction products was only ~14 μm --or about 0.2% of the total liner thickness. Maximum penetration was 40 μm . At the accelerated reaction rate, the time required to penetrate the entire depth of the liner might be increased by a factor of 200 to 500. Based on the testing that represented seven years of aging, the asphalt membrane performance could be expected to greatly exceed the 1000-year standard. If needed to prevent groundwater contamination, catalytically airblown asphalt could, therefore, be expected to perform extremely well as a leachate barrier at inactive uranium mill tailings piles for long periods.

CONTROLLED CLAY LINER EVALUATIONS

Although the sodium bentonite amended to natural soil proved to be the most effective leachate barrier of the clay liner category, natural soils without amendments were considered to have adequate retardation capabilities for most inactive mill tailings sites. Therefore, in addition to conducting further studies with sodium bentonite amendments, two calcareous soils from Clive, Utah, were evaluated.

These evaluations were conducted not only to determine permeability but also to assess the sorptive and coprecipitative properties of the clay and bentonite amended soils. After material characterizations, the soils and clay liner were interacted with acidic and alkaline leachates; and batch and column interaction studies were performed. The batch studies indicated overall retardation capabilities, such as sorption and buffering, while the column studies determined permeability and rate of contaminant breakthrough.

Material Characterization

The bentonite liner material (soil plus 10% sodium bentonite) and two soil samples from Clive, Utah (Soil #2 and Soil #6) were characterized (see Table 13). The soils from Clive had a very high calcium carbonate (CaCO_3) content, and an unusually high level of excess salts (compared with normal

TABLE 13. Characteristics of Clive, Utah, Soil Samples

Depth	Soil #6	Liner Material (Soil plus 10% Na-Bentonite)	Soil #2
	30-61 cm	30-61 cm	180 cm
<u>Particle Size</u>			
% Sand	17	15	13
% Silt	46	42	31
% Clay	36	42	55
% Organic Matter	0.91	0.93	0.66
<u>Carbonate Content</u>			
% CaCO ₃	27	25	57
<u>Cation Exchange Capacity</u>			
(meq/100 g)	18	24	15
<u>Surface Area</u> (m ² /g)	77 ±15	131 ±8	62 ±13
<u>Saturated Paste</u>			
pH	8.3	8.4	8.6
E.C. (mmhos/cm)	48	30	14
<u>Buffering Capacity</u> ^(a)			
HCl equivalent (meq/100 g)	450	410	1100

(a) Amount of total acidity required to lower the soil pH to 4.5.

agricultural soils) was indicated by the high electrical conductivity measurements made on saturated soil pastes. Ionic strengths were calculated using the method described by Lindsay (1979). Saturated paste E.C. measurements indicated that ionic strength ranged from 0.2 to >0.7 moles/L.

To determine buffering capacities, 1 g of soil in 50 cm³ of distilled water was titrated with HCl. During titration, the pH dropped very slowly between pH 8 and pH 7, but more quickly between pH 7 and pH 5. Below pH 5, however, the value dropped rapidly to <2.0.

The mineralogical compositions of these soils and liner material were analyzed by X-ray diffraction on random-packed powder mounts and oriented slurries of each bulk sample. Based on the relative intensities of certain reflections, semiquantitative estimates were made of the relative abundance of mineral phases.

Both soils and the liner material (soil plus 10% sodium bentonite) contained quartz, carbonates (calcite, dolomite and aragonite), kaolinite, illite and feldspar. The carbonates and quartz were the major phases, and the clay minerals (kaolinite and illite) were minor phases. Soil #2 contained the least feldspar and the most aragonite.

Leachate Interactions with Soil and Liner Materials

Batch and column methods were both used to study the interactions of tailings leachates with the two soils and the liner material. Synthetic leachates were prepared to represent the initial 0.20 to 0.30 pore volumes of effluent from the tailings column leach tests (see Table 14). For the batch tests, 250 cm³ of synthetic tailings leachate were shaken with 50 g of soil or liner material in a 1-L polyethylene bottle (similar to the 5:1 batch tailings leach tests).

The soils and liner material were titrated with synthetic leachate, the same procedure used to measure the buffering capacity except that synthetic leachate was substituted for HCl.

Permeability values (saturated hydraulic conductivities) were measured using columns that were 8.9 cm long and 8.9 cm in diameter. Synthetic leachate was percolated upward through each column and collected in separate covered graduated cylinders. Pressure was maintained on the leachate using compressed air in the leachate reservoir.

Titration experiments and batch contact of synthetic leachate with the soils and liner material have shown that these earthen materials are very effective at neutralizing acidic leachates but not the alkaline Durango leachate.

TABLE 14. Comparison of Synthetic Leachate Compositions Before and After Contact with Soil #6 in 5:1 Solution to Solid Ratio Batch Test (concentrations are in mg/L)

	Shiprock		Salt Lake		Durango	
	Before	After	Before	After	Before	After
Al	2860	0.27	1230	0.12	5.3	0.24
Ca	521	420	1.1	500	5.6	3.0
Cu	130	0.06	47	0.02	ND	0.02
Fe	177	0.09	1800	0.04	0.04	0.01
K	61	160	ND	21	44	72
Mg	2060	2000	165	250	16	10
Mn	67	19	56	0.4	0.16	0.07
Na	1560	2600	119	1200	2080	2500
Ni	11	ND ^(a)	22	ND	ND	ND
Sr	1.9	3.3	ND	7.1	0.06	0.06
Zn	112	0.2	86	0.1	ND	0.1
B	ND	4.7	ND	4.9	0.02	3.2
Cl	874	3600	1110	1300	26	870
CO ₃	ND	600	ND	200	1800	1800
F	16	9.7	3.5	ND	17	1.8
Mo	ND	0.2	ND	0.03	0.8	0.1
NO ₃ + NO ₂	870	130	307	ND	74	60
PO ₄	14	ND	6	ND	76	45
SO ₄	30,000	8500	12,500	2600	1370	290
Si	11.5	2.2	ND	1.5	2.1	4.5
As (ppm)	3.8	0.2	0.1	≤0.1	1.2	0.7
Ba	ND	0.01	ND	0.09	0.1	0.02
Cd	4.7	0.05	2.5	≤0.01	0.01	≤0.01
Cr	1.9	≤0.02	2.3	≤0.02	0.07	0.04
Pb ^(b)	5.0	0.2	2.4	0.2	ND	ND
pH	2.1	7.7	2.1	7.7	9.7	9.4
E.C. (mmho/cm)	26	20	11.0	8.2	10.0	9.5
Percent Sodium ^(c)		38		53		97
Total Cations (meq/L) ^(d)	600	300	227	98	94	112

(a) ND = Not detected.

(b) 5 ppm Pb added to the synthetic leachate.

(c) Percent sodium = $100 \times [\text{Na}] / (\text{Total cations}) \text{ meq/L}$.

(d) Total cations = the sum of all cations in meq/L.

TABLE 15. Buffering Capacities of Clive, Utah, Soils and Liner Material

	<u>Soil #6</u>	<u>Liner Material (Soil plus 10% Na-bentonite)</u>	<u>Soil #2</u>
<u>Carbonate Content</u>			
Weight % CaCO_3	27	25	57
CaCO_3 (meq/100 g)	540	500	1140
<u>HCl Equivalent</u>			
(meq/100 g) ^(b)	450	410	1100
<u>Leachate Equivalent</u>			
(meq/100 g) ^(b)	470	420	1250

(a) Amount of acid (HCl) required to lower the soil pH to 4.5.

(b) Total acidity (as leachate) required to lower the soil pH to 4.5.

The soils described in Table 13 were titrated with synthetic Shiprock tailings leachate for comparisons with the buffering capacities measured by HCl titration and those calculated from CaCO_3 contents. Total acidity for the Shiprock leachate was calculated using the sum of Al^{+3} , Fe^{+3} and H^+ concentrations in milliequivalents per liter. The buffering capacity measurements (to pH 4.5) listed in Table 15 are very close to the CaCO_3 contents in these soils.

Below pH 4.5, a significant difference was observed between the titration curves produced by Shiprock leachate and HCl. The HCl titration indicates the buffering capacity of the soil carbonates. The pH dropped slowly to 4.5; then, addition of more HCl caused a rapid decline in pH to less than 2.0. Figure 23 shows a very large buffering capacity below pH 4.5 when Shiprock leachate was used as the titrant but not when HCl was used.

Titration of distilled water with Shiprock leachate produced a curve similar to the soil-leachate curve when the pH dropped below pH 4.5. A

FIGURE 23. Titration Curves for Soil #2 Using HCl and Shiprock Tailings Leachate (dashed line)

cloudy suspension resulting from the blank titration also indicated that a precipitation reaction had occurred. Because of the relatively high solubility of Fe under the conditions of the blank titration, it is likely that Al^{+3} precipitated. Dissolution of the Al precipitate below pH 4.5 seems to be the most likely mechanism for the "extra buffering capacity" of this soil for Shiprock tailings leachate (see discussion below).

The information presented above indicates that soils from the Clive, Utah, site have a very high buffering capacity for acidic tailings leachate. The total acidity of tailings leachate equals the sum of the concentrations of H^{+} , Al^{+3} and Fe^{+3} (meq/L). For the Shiprock tailings leachate, more than 95% of the total acidity was caused by hydrolysis of Al^{+3} as the leachate pH increased on contact with the soil. Several Al compounds such as gibbsite and alunite may also precipitate but only amorphous $Al(OH)_3$ will be considered for simplicity. The hydrolysis of Al^{+3} can be expressed as

As Al^{+3} hydrolyzes, it releases H^{+} to solution which can react with solid carbonate phases that neutralize the acidity

As the CaCO_3 dissolves and releases Ca^{2+} , the high sulfate concentrations present in the leachate cause precipitation of gypsum:

When the pH drops to about 4.5, the pH buffering capacity of the carbonates is exhausted; however, Al compounds may buffer the pH through the reversal of the Al precipitation reaction

As the Al compounds dissolve, H^{+} is consumed and the pH is buffered below 4.5 as shown in Figure 23.

These reactions have three effects: 1) neutralization of leachate acidity to pH 4.5 [see Figure 23 (equivalent to the soil CaCO_3 content)]; 2) increase in the solid-phase volume because of the lower specific gravity of gypsum (2.3 g/cm^3) compared to soil carbonates (2.7 to 3.0 g/cm^3); and 3) precipitation of Fe and Al compounds that may buffer the leachate pH below pH 4.5.

The precipitation of gypsum, aluminum and iron compounds and precipitates of other cations from the neutralized leachate will reduce contaminant levels through adsorption/coprecipitation reactions and reduce the porosity and permeability of the soil or liner material.

Analysis of the end point (pH 3.8) solution resulting from the soil titration with synthetic Shiprock leachate showed that Cd, Ce, Li, Mg, Mn, Na, and Zn concentrations decreased by 50-70%, while Al, As, Cr, Cu, Fe, and SO_4 concentrations decreased by >70% compared to the initial leachate

concentrations. Dilution of the leachate during titration would account for only a 33% decrease in concentrations. The rest of the concentration decreases were caused by precipitation, coprecipitation, and adsorption reactions.

Table 14 lists chemical compositions of the three leachates after contacting Soil #6. (These results are similar to the leachate compositions after contact with the other two soils). Precipitation caused total cation (and anion) concentrations to decrease in both acidic leachates.

Analysis of soil mineralogy by X-ray diffraction after contacting acidic leachate from Salt Lake and Shiprock tailings indicated that 1) soil carbonates (calcite, dolomite, and aragonite) decreased in abundance, and 2) gypsum formed in the soils. Gypsum was not detected in soils contacting alkaline Durango leachate (although sulfate concentrations decreased significantly in the leachate), and the soil carbonate abundances were apparently unchanged by contact with Durango leachate.

Permeability values for the liner material (soil plus 10% sodium bentonite) decreased with time as leachate passed through the columns. Sodium-saturated bentonite clay is known to swell and disperse when contacted with low ionic strength solutions having a high percentage of dissolved sodium (Sherard, Dunnigan and Decker 1976; Cass and Sumner 1982). At high ionic strengths, the clay may remain flocculated even when the solution has a high sodium percentage.

The soils and liner studied contained a considerable amount of excess sodium salts, which appeared to prevent dispersion and swelling of the clay upon initial contact between leachate and liner. The liner permeability generally decreased as the effluent electrical conductivity (therefore ionic strength) decreased. This is a good indication that the sodium bentonite became dispersed and swelled as the excess sodium salts were washed from the column. The dispersed and swollen clay particles plugged up conducting pores in the liner and reduced the permeability.

FIGURE 24. Saturated Hydraulic Conductivity K , cm/s, as a Function of Durango Tailings Leachate Pore Volumes Passing Through the Soil Columns

Permeability values for Durango leachate (see Figure 24) contacting the two soils without bentonite tended to remain constant with time even though the electrical conductivity (ionic strength) decreases; however, permeability of the bentonite-amended soil decreased. The Durango leachate had a low ionic strength (compared to the two acid leachates) and a sodium percentage of 96%, which would disperse the bentonite, according to Sherard et al. (1976). The constant permeability values for the other two soils that contacted Durango leachate support the argument that dispersion and swelling of the clay contributed to the decrease in permeability observed for the liner material.

For the alkaline Durango leachate, clay swelling and dispersion were probably the major mechanisms for a decrease in permeability. For the two acidic leachates, however, precipitation of gypsum and other compounds may also contribute to observed decreases in permeability.

FIGURE 25. Saturated Hydraulic Conductivity K , cm/s, as a Function of Shiprock Tailings Leachate Pore Volume Passing Through the Soil Columns

Experimentally observed permeability values decreased to about 10^{-10} cm/s or less for all soil or liner columns contacting the high sulfate (34 grams of sulfate per liter) Shiprock leachate (Figure 25). Permeability values for the lower sulfate (13 g/L) Salt Lake leachate contacting the high carbonate soil (55% CaCO_3 equivalent) and the liner material (25% CaCO_3 with 10% bentonite) both decreased to $<10^{-10}$ cm per second. However, the combination of low sulfate leachate (Salt Lake) and low carbonate soil (25% CaCO_3) without bentonite resulted in a relatively constant permeability of about 10^{-9} cm/s (Figure 26).

The Salt Lake leachate had less total acidity (50%), less dissolved Al (50%), and less sulfate (40%) than the Shiprock leachate. Thus, precipitation of gypsum, Al, and Fe compounds from Salt Lake leachate would be not be expected to occur as rapidly (at the same flow rate) as it would from Shiprock leachate. If all CaCO_3 in Soil #6 (25% CaCO_3) dissolves, gypsum, Al, and Fe precipitates will form and a decrease in both porosity and permeability should eventually be observed after more pore volumes of leachate have percolated through the column.

FIGURE 26. Saturated Hydraulic Conductivity K , cm/s, as a Function of Salt Lake Leachate Pore Volumes Passing Through the Soil Columns

Characterization of the tailings showed that both Shiprock and Salt Lake tailings leachate were in equilibrium with gypsum to at least two pore volumes. Therefore, gypsum precipitated during neutralization of leachate acidity by soil carbonates should not dissolve on contact with fresh (un-neutralized) leachate as long as leachate equilibrium with gypsum is maintained (i.e., high Ca concentrations). Moreover, the permeabilities of these soils or liners will not increase later as a result of acid leachate dissolution of gypsum.

In summary, these studies demonstrated that the pH buffering capacities of the two Clive, Utah, soils and one liner material for acidic tailings leachate resulted from two reactions: 1) dissolution of soil carbonate phases, and 2) dissolution of Al (and perhaps Fe) precipitates formed during the leachate-carbonate reaction.

Permeability values will decrease with time under at least two sets of conditions: 1) contact between low ionic strength, high-sodium percentage leachates and liners or soils rich in sodium-saturated bentonite that causes the clay to disperse and/or swell and plug up pore spaces; and 2) contact between high sulfate acidic leachate and soil or liners with high carbonate contents that cause gypsum precipitates and plug up pore spaces, reducing porosity.

FIELD LINER EVALUATIONS

The catalytic airblown asphalt membrane and natural soil amended with sodium bentonite were also subjected to field studies beginning in September 1981. The liners were installed in a pit carved out of the Grand Junction tailings pile, instrumented, and covered with tailings, which were subsequently saturated with water. After an exposure of almost 2 years, the materials were sampled and analyzed for performance.

TEST DESCRIPTION

Figure 27 illustrates the construction of the pit in which the liners were installed. The pit was 29 m (96 ft) long, 22 m (72 ft) wide, and 1.8 m (6 ft) deep. The slope of the walls was limited to an approximate 4:1 grade. To isolate any extraneous moisture and prevent its migration into the foundation soil from the tailings beneath the liners, a moisture barrier consisting of clear rock, 25-35 cm (10-14 in.) thick, was placed on top of the tailings. This provided a capillary moisture barrier so that any moisture accumulated in the foundation soil beneath the liners could be attributed to permeation through the liners.

FIGURE 27. Cross Section of Tailings Pit for Liner Evaluation in the Field

The clear rock layer was covered with a thin sheet of polyethylene membrane to keep the foundation soil from sifting into the voids. The membrane was not installed to prevent moisture migration downward; thus, as portions of the foundation soil became saturated, they could drain through the clear rock. A 25-35 cm (10-14 in.) thick layer of foundation soil (overburden) was installed over the polyethylene membrane.

The foundation soils underlying the asphalt membrane and sodium bentonite liners were separated by the polyethylene membrane at the centerline of the pit. This prevented cross-contamination of leachates that might have permeated either liner and allowed each liner to be evaluated independently.

The foundation soil under each liner was instrumented with an array of moisture blocks and moisture extraction cups (see Figure 28 for locations). The moisture blocks provided readings on the change of saturation profiles in the foundation soil, and the moisture extraction cups provided samples of leachate that passed through the liner(s) even under unsaturated foundation soil conditions.

The catalytic airblown asphalt membrane (specifications are listed in Appendix B) was sprayed at 200°C from a distributor truck over the western half of the pit. Approximately 2600 kg of catalytic airblown asphalt was sprayed to a thickness of 0.7 cm in two passes. Figure 29 is a photograph of the application. The moisture extraction cup tubes, through which leachate samples were withdrawn from beneath the liner, protruded through the asphalt membranes.

On the eastern half of the pit, 6800 kg of Wyoming-quality sodium bentonite was spread at a concentration of 20 kg/m² over the 30-cm layer of foundation soil. It was rototilled into the soil to a depth of 10 cm (shown in Figure 30), moistened to 12-18% moisture along the walls of the pit, and compacted. Great care was taken to maintain a good seal between the bentonite layer and moisture extraction cup tubes, which protruded through the liner.

FIGURE 28. Locations of Moisture Blocks and Extraction Cups

Following inspection of the liner installations, tailings were reintroduced into the pit and saturated with fresh water. Piezometers were installed in the tailings at the same locations as the moisture extraction cups to determine the saturated water level. The tailings were covered with a layer of overburden to prevent airborne contamination by windswept tailings.

MOISTURE BLOCK DATA

The moisture blocks were buried a few centimeters below the bottom of each liner. These instruments measure the degree of saturation of the foundation soil, which is equal in percentage to the saturation ratio. These values indicated the effectiveness of each liner as a leachate barrier. Foundation soil moisture contents at various times after installation of the liner materials are shown in Figure 31.

FIGURE 29. Spray Application of Asphalt Membrane

FIGURE 30. Rototilling of Bentonite into Foundation Soil

FIGURE 31. Degree of Saturation of Foundation Soil Below the Liners at Selected Times

The saturated foundation soil condition beneath a portion of the asphalt membrane shortly after installation (September 1981) was caused by misplacement of the polyethylene divider between the foundation soils. Rainfall over the eastern half of the pit during installation of the bentonite liner, and leachate that subsequently migrated through the bentonite liner, moved horizontally through the foundation soil beneath the asphalt membrane. The misplaced polyethylene divider was reinstalled in November 1981 and, as can be seen by the isograms, the foundation soil moisture content began to decrease slowly. Except for a small area near the center, data collected after that time showed that the asphalt membrane had been an effective barrier to leachate migration.

A leak was detected in the center of the pit where an inadequate thickness of asphalt occurred in a one-square-meter area. This was discovered when the liner was partially uncovered and sampled in May 1983. The thin portion of the liner had small-diameter (1-cm) holes that allowed leachate to pass slowly.

On the other hand, the clay liner did not demonstrate the same type of effectiveness as a leachate barrier. It was not expected to be as impermeable, especially for the near-neutral tailings conditions found at Grand Junction. Although leachates were able to migrate through the clay liner, it was an effective cation exchanger for adsorbing contaminants from the leachates as they passed through. The performance of the clay liner will be discussed in more detail in the following section, which presents data from leachate samples collected above and below the liner.

Figure 32 compares the saturation ratios for selected locations under the asphalt and clay liners. As the figure shows, the degree of saturation under the clay liners rose to almost 100% in most cases within a few months of saturating the tailings above the liner. Saturation of the foundation soil beneath the asphalt membrane, however, remained relatively constant following the repair of the polyethylene divider.

FIGURE 32. Degree of Saturation Under Asphalt and Clay Liners at Selected Locations

Piezometers indicated the water levels in the tailings during this period (see Figure 33). The rapid decrease in water level above the clay liner was caused by evaporation through the cover and leachate permeation through liner. Since the asphalt membrane was much more effective as a leachate barrier, the gradual decrease in water level for the tailings above the membrane must be attributed primarily to evaporation through the cover. In addition, a small portion would have leaked through the 1-cm holes caused by faulty installation in a small area of the liner.

FIGURE 33. Water Level in Tailings Above Asphalt and Clay Liners

LEACHATE SAMPLE ANALYSIS

Leachate samples were collected from the piezometers in the tailings above the asphalt and clay liners on a monthly basis as well as from the moisture extraction cups beneath the liners when conditions permitted. Analyses of the samples showed that the clay liner was effective as a barrier to contaminants identified in the Environmental Protection Agency (EPA) Drinking Water Standards (U.S. EPA 40 CFR 141).

The results of the piezometer and moisture extraction cup sample analyses are listed in Table 16. The samples were analyzed for elemental As, Ba, Cd, Mo, Se, Ag, and Hg. Chloride, nitrate, and sulfate anions were also analyzed. The tabular data indicates that the clay liner effectively

TABLE 16. Leachate Sample Analysis

Sample Description	Element	Standard Number of Samples	EPA Mean ($\mu\text{g/mL}$)	Deviation ($\mu\text{g/mL}$)	Standard ($\mu\text{g/mL}$)	Reduction Factor
LEACHATE FROM PIEZOMETERS ABOVE BOTH LINERS						
	As	9	1.86	0.85	0.05	--
	Ba		0.23	0.10	1.00	--
	Cd		<0.01	0	0.01	--
	Mo		10.9	2.1	0.05	--
Se		2.9	0.7	0.01	--	
Ag		0.09	0.06	0.05	--	
Hg		<0.014	0.013	0.002	--	
Cl ⁻		2.86	1.96	--	--	
NO ₃ ⁻		1.08	0.84	10	--	
SO ₄ ⁻²		2.24	0.29	--	--	
pH		7.88	0.40	--	--	
LEACHATE FROM MOISTURE EXTRACTION CUP BENEATH CLAY/SOIL LINER						
	As	2	0.045	0.05	0.05	41.3
	Ba		0.095	0.007	1.00	2.4
	Cd		<0.01	0	0.01	--
	Mo		0.70	0.14	0.05	15.6
	Se		0.55	0.071	0.01	5.3
	Ag		0.07	0.014	0.05	1.3
Hg		<0.01	0	0.002	--	
Cl ⁻		3.5	0.28	--	0.82	
NO ₃ ⁻		6.4	0.78	10	0.17	
SO ₄ ⁻²		6.4	0.42	--	0.36	
pH		8.11	0.10	--	--	
LEACHATE FROM MOISTURE CUP BENEATH ASPHALT MEMBRANE						
	As	3	0.07	0.08	0.05	27
	Ba		0.12	0.08	1.00	1.9
	Cd		<0.01	0	0.01	--
	Mo		4.1	3.1	0.05	2.7
	Se		1.5	1.1	0.01	1.9
	Ag		0.04	0.03	0.05	2.2
Hg		<0.01	0	0.002	--	
Cl ⁻		3.2	1.1	--	0.89	
NO ₃ ⁻		1.9	0.8	10	0.57	
SO ₄ ⁻²		2.7	0.3	--	0.83	
pH		8.18	0.12	--	--	

reduced the elemental contaminant concentrations either by factors of 5-50 or to within the drinking water standard.

In the case of Cd, the concentration in the leachate above the liner was below both the analytical detection limit and the standard. Silver reduction was only a factor of 1.3, but the arithmetic mean of Ag concentrations in the leachate samples below the clay liner was very near the drinking water standard of 0.05 $\mu\text{g/ml}$. In all cases, mercury concentration was below the detection limit of 0.01 $\mu\text{g per/mL}$. The leachate concentration of Ba below the clay liner was 2.4 times less than that in the leachate above the liner; however, the concentrations below and above the liner were both below the MCL.

The reduced contaminant concentration is attributed primarily to the sorption of cations onto the sodium bentonite clay minerals. Coprecipitation of contaminants in the leachate was not expected to be a factor in this case because the pH of the piezometer leachate samples and the moisture extraction cup samples were both near neutral (i.e., between 7.13 and 8.25 pH).

Precipitation of cations was not as significant as it would have been for an acidic leachate contacting this soil/clay liner; however, As, Mo, and Se concentrations were reduced. These reductions were not the result of cation exchange because As, Mo, and Se most likely exist as anionic species in these leachates. Anion adsorption is not likely to be important either because of the high pH. The only apparent mechanism left to explain the reduction of As, Mo, and Se concentrations is their precipitation in as-yet unidentified compounds. By the process of elimination, therefore, precipitation reactions for some elements do contribute to the effectiveness of soil and clay liners for neutral leachates.

No reduction in contaminant concentrations was observed for chlorides, nitrates, and sulfates. The average chloride concentrations of the piezometer and extraction cup leachates were within each other's standard deviation; thus, the clay liner was not sorbing any of the chloride ions. On

the other hand, the nitrate and sulfate concentrations below the clay liner were on the order of 3-6 times those of the concentrations in the piezometer leachate--probably due to interactions with the foundation soil as the leachate passed through the liner. The concentrations of sulfates, nitrates, and chlorides in the foundation soil were inconsequential, however, because they were either below the EPA drinking water MCL's or not listed. In the case of higher sulfate concentrations in the leachate, which would occur in primarily acidic tailings, the sulfate concentration would be reduced by precipitation of sulfate compounds as the leachates were neutralized while passing through the clay/soil liner.

Table 16 also lists the leachate concentrations below the asphalt liner. These data show that the concentration of contaminants has not been reduced in the leachate below the asphalt liner to the same degree as the clay liner. The only leachates collected under the asphalt liner were near the leaking section described earlier. The reduction in contaminant concentrations, especially that of arsenic, was probably due to the interaction of the leachate with the foundation soil. Even so, the contaminant reduction was not as pronounced as those leachates that had permeated the clay liner. Thus, the foundation soil alone was apparently not as effective as the combination of bentonite and soil in sorbing and precipitating the clay contaminants from Grand Junction leachates.

ASPHALT LINER EVALUATIONS

In May 1983, the liners were partially uncovered for sampling. In the west end of the pit, three asphalt liner samples were collected from three separate locations. The samples were analyzed and compared with a laboratory aged sample and an unexposed sample that was taken from the field membrane in September 1981.

A procedure developed by Geir and Morrison (1968) was used to make the following determinations:

- statistical variation of membrane thickness
- thickness of crust formation on the exposed surface
- extent of soil impregnation from the underlying soil.

One of the locations was selected specifically to determine the cause for the high saturation ratios detected under a portion of the liner. It was discovered that, due to improper application and inspection practices, a one-square-meter portion of the liner had been applied with insufficient asphalt. This resulted in holes approximately 1 cm in diameter.

According to the Geir and Morrison procedure, this sample received a fair to poor rating, primarily because of its discontinuity. The other two samples, which had very few surface cracks, a thin layer of crust, good flexibility, and no physical damage, earned good ratings.

Table 17 compares the three field samples and the laboratory-aged and virgin samples. The table shows that the two properly installed field samples compared very well with the laboratory-aged samples, supporting the conclusion that the aging tests were successful in accelerating the degradation reactions in the laboratory.

TABLE 17. Comparison of Field, Virgin, and Laboratory-Aged Asphalt Liners

	Field Liner			Virgin Sample	Laboratory-Aged Sample	
	No. 1	No. 2	No. 3		Liner A	Liner 4
General Appearance (a)	Good	Fair to Poor	Very good	Excellent	Good	Good
Crust Thickness (% of total)	5-8	8-10	1-2	0	2	2
Soil Impregnation (% of total thickness)	10	20-100	20-50	5-20	30	30

(a) Good is defined as few surface cracks, thin layer of crust, flexible, and small amount of physical damage. Fair is defined as some surface cracking with a few large cracks, a portion of the surface crust brittle, some foreign material incorporated, and some physical damage. Poor is defined as brittle and lifeless, large amount of foreign material incorporated, and extensive physical damage (Geir and Morrison 1968).

The uniformity of thickness of the three field samples was also measured at 10 random locations, each to determine the adequacy of application. The average thicknesses for liner samples 1, 2, and 3 were 6.33 mm (standard deviation = 2.43), 4.24 mm (standard deviation = 1.46 mm), and 7.17 mm (standard deviation = 1.94 mm), respectively. Only liner 2, which was inappropriately applied, was significantly below the design thickness of 7 mm.

The experience with the formation of an inferior portion of the liner during installation demonstrated the need for appropriate design and inspection practices. Recommended construction procedures for buried asphalt membranes are outlined by Geir and Morrison (1968) and Van Asbeck (1964) and are summarized below.

- The maximum preferred steepness of a side slope is two horizontal to one vertical. The limiting factor is stability of the foundation soil and preparation and application equipment. The demonstrated slope thickness during the field tests was 4:1, which should be attained if at all practicable.
- The foundation soil must be dragged to remove sharp objects and expose rocks for removal.
- The foundation soil should be rolled with a smooth steel roller to minimize soil impregnation and the amount of asphalt required to obtain a uniform thickness.
- If the subgrade is dusty, a light sprinkling of water is necessary. Failure to do this in the 1981 field tests resulted in a 10-50% soil impregnation. Although a thick coat of cationic emulsion prior to membrane application has been used in other field tests, this is not recommended because it does not allow access by the asphalt distributor truck.
- The asphalt must meet the specifications outlined in Table B.3 (Appendix B) and use a P_{205} catalyst during the manufacturing process.

- The asphalt must be applied at 204°C with a slot-type spray bar in a minimum of two and preferably three passes. Multiple passes reduce soil impregnation and reseal pinholes from escaping moisture.
- Recommended application speed is 6.5 km/h.
- Visual inspection of the entire surface is necessary.
- If tailings are not added to the pit immediately after application, a 30-cm thick protective soil cover is recommended to protect the membrane from sunlight, high temperatures, heavy equipment, and vectors.

CLAY LINER EVALUATION

Six core samples were removed from the clay liner field test pit in May 1983. The cores were analyzed to determine the effects of field conditions on the chemical and physical properties of the test liner. All cores were collected in clear lexand tubing so that core quality could be monitored in the field prior to shipment back to the laboratory for analysis.

Three cores contained only the clay liner portions of the overall liner system and measured 8.10 cm in length and 8.86 cm in diameter. Each of the remaining three cores, which measured 30.5 cm in length and 8.86 cm in diameter, contained material from the three layers of the liner test system: leached tailings, clay liner, and foundation soil.

Bulk tailings samples contained leached tailings from directly above the clay liner and undisturbed tailings collected adjacent to the test pit. In addition, a sample of laboratory-prepared clay liner was produced from nine parts uncontacted foundation soil and one part Wyoming-grade sodium bentonite. A brief description of each core is provided in Table 18.

TABLE 18. Field Sample Description

<u>Sample Number</u>	<u>Core Length and Description</u>
Core No. 1	8.1-cm clay liner core
Core No. 2	8.1-cm clay liner core
Core No. 3	8.1-cm clay liner core
Core No. 4	30.48-cm core containing leached tailings, clay liner, foundation soil
Core No. 5	30.48-cm core containing leached tailings, clay liner, foundation soil
Core No. 6	30.48-cm core containing leached tailings, clay liner, foundation soil
Leached Tailings	Leached tailings collected from above the clay liner from sampling hole No. 4
Fresh Tailings	Uncontacted tailings collected adjacent to the liner site

Each column was subsampled into five equal lengths:

Column top - 24.4 cm

24.4 cm - 18.3 cm

18.3 cm - 12.2 cm

12.2 cm - 6.1 cm

6.1 cm - column bottom.

The average depth of the clay liner from the column top was 22 cm. The first two subsamples (column top - 24.4 cm and 24.4 cm - 18.3 cm) contain mostly leached tailings. Assuming a clay liner thickness of 10 cm, the third subsample (18.3 cm - 12.2 cm) was predominantly clay liner. The last two subsamples contained foundation soil.

Sediment Analysis

The optimum physical properties of the clay liner were tested using laboratory-produced liner material, which was tested for optimal proctor density and particle size distribution by standard ASTM methods. For comparison, contacted clay liner material obtained from Core No. 3 was also tested for field water content, dry bulk density, and particle size distribution. Results of these tests indicated the effectiveness of preparing

and installing a clay liner under field conditions. Measurements of hydraulic conductivity were made using Cores No. 1 and No. 2, by a modified ASTM falling head permeability method (ASTM 1978). Flow through the cores was from bottom to top to ensure saturated conditions.

Solution Analysis

Solutions were prepared from every sediment sample by the method of 1:1 extraction with distilled water. Extracts were produced by adding a known weight of sediment to an equal weight of distilled water. The resulting slurry was placed in 0.5-L polyethylene bottles and shaken for 48 hours on a linear oscillating shaker to facilitate mixing. Upon completion of the two-day contact period, samples were centrifuged and supernates were filtered through a 0.45-micron membrane. Each 1:1 extract sample was split into four aliquots for analysis. Ten milliliters were sent for pH and Eh determination, 10 to cation analysis, 10 to anion analysis, and at least 200 to radionuclide analysis.

Radionuclide concentrations were determined by high-resolution gamma spectroscopy. For this analysis, a known volume of solution (at least 200 mL) was evaporated. The resulting evaporates were mixed with a cellulose binder and pressed into a pellet of fixed geometry. A Princeton Gamma-Tech intrinsic germanium diode coupled with a Nuclear Data ND660 nuclear analyzer was used for all counting.

RESULTS AND DISCUSSION

PHYSICAL CHARACTERISTICS

The results of proctor testing and particle size analysis for the laboratory-prepared clay liner are presented in Figures 34 and 35, respectively. Particle size distribution of the contacted field liner obtained from Core No. 3 is given in Figure 36. For comparison, the water content, dry bulk density, and particle size distribution data for both samples are presented in Table 19.

The results listed in Table 19 indicate that field preparation and installation of the test clay liner were very successful. Although the field liner carries a soil classification of loam and the laboratory-prepared liner a classification of silt loam, particle size distribution data showed the two mixes to be very similar. Under the unified soil classification system, both liner materials were classified as fine-grained inorganic silts (ML). This indicated that a properly proportioned mixture was achieved under field conditions.

In addition, the dry bulk density of the field test liner was within 88% of proctor, as determined by testing of the laboratory-prepared liner. Along with a field water content of 20.4 weight percent, the proctor data indicated that the liner could be compacted to, and maintain, even higher bulk density values in the field. An increase in field bulk density would result in decreased liner permeability and increased liner resistance to leachate migration.

Hydraulic conductivity data obtained from Cores No. 1 and 2 showed permeability ranging from 1×10^{-7} cm/s to 5×10^{-9} cm/s, respectively. Dry bulk density calculations for each core placed Core No. 1 at 93% of proctor (bulk density = 1.64 g/cm^3) and Core No. 2 at 100% of proctor (bulk density = 1.87 g/cm^3). Bulk density and water content analyses of the subsamples removed from Cores 4, 5, and 6 showed bulk densities ranging from 85-95% proctor and water contents between 20 and 22 weight percent.

FIGURE 34. Moisture Density Relationship of Laboratory Prepared Clay Liner

FIGURE 35. Particle Size Distribution of Laboratory Prepared Clay Liner

FIGURE 36. Particle Size Distribution of Field Test Clay Liner

TABLE 19. Physical Characteristics of Field and Laboratory Clay Liners

Parameter	Core No. 3 Clay Liner Material	Laboratory Produced Clay Liner
Moisture Content (wt% as received)	20.35	17.61
Particle Density (g/cm^3)	2.74	2.76
Dry Bulk Density (g/cm^3)	1.56	1.77 ^(a)
Particle Size Analysis <-----% less than---->		
2,000 μm	99.93	98.60
500 μm	99.90	97.02
250 μm	99.28	95.13
106 μm	89.08	89.09
53 μm	73.09	72.61
9 μm	35.67	29.15
1.3 μm	21.33	15.26
USDA Soil Classification	Loam	Silt Loam

(a) Values represent soil at 100% Proctor.

Clay Liner Attenuation Characteristics

Sherwood and Serne (1983) reported that the mobility of many dissolved constituents, including cations, trace metals and radionuclides, in acidic uranium mill tailings leachates is controlled by solution pH. When acidic uranium mill tailings leachates were neutralized to pH = 7.0 or above, average removal percentages greater than 98% were reported for such contaminants as Al, As, Ba, Cr, Co, Cu, Pb, Si, and ^{210}Pb and ^{230}Th . These results indicate that such percentage reductions can be achieved regardless of the neutralizing agent employed. Leaching studies conducted by Serne, Peterson, and Gee (1983) indicated that when acidic mill tailings effluents were leached through sediment materials containing high buffering capacities, effluent quality, as indexed by low concentration of radionuclides and heavy metals, remained high as long as the effluent pH was above pH = 4.0.

As a first check of the potential level of contaminant migration through the Grand Junction test clay liner, pH was monitored with depth through the liner system. The results of this investigation are found in Table 20, which also includes the pH of uncontacted and leached tailings. Table 20 presents pH and Eh values to represent saturated paste values of subsamples from Core No. 5.

TABLE 20. Core No. 5 Saturated Paste pH, Eh, and Ec Data with Respect to Depth

<u>Core Depth</u>	<u>pH (units)</u>	<u>Eh (mV)</u>
Fresh Tailings	7.39	305
Leached Tailings	7.98	294
Column Top - 24.38 cm	8.51	259
24.38 cm - 18.29 cm	7.34	335
18.29 cm - 12.19 cm	8.01	265
12.19 cm - 6.01 cm	8.36	283
6.01 cm - Column Bottom	7.73	256

TABLE 21. 1:1 Extract pH, Eh, and Ec Data with Respect to Depth

<u>Core Depth</u>	<u>pH (units)</u>	<u>Eh (mV)</u>
Fresh Tailings	7.87	301
Leached Tailings	8.28	291
Column Top - 24:38 cm	8.05	260
24.38 cm - 18.29 cm	7.92	282
18.29 cm - 12.19 cm	7.89	268
12.19 cm - 6.01 cm	8.18	278
6.01 cm - Column Bottom	7.60	277

Results of pH, Eh, and Ec data taken for 1:1 extract solutions are given in Table 21. The pH and Eh values are average 1:1 extraction values obtained from the analysis of 1:1 extracts of subsamples of Cores 4, 5, and 6.

From the pH data presented for the test liner system, it is evident that neutral pH conditions exist throughout the liner system. Thus, low leachate concentrations can be expected of all contaminant species displaying solubility restrictions at neutral pH. Because several of the contaminating constituents of the leachate source and the clay liner material will be insoluble at the test liner system's neutral pH, X-ray fluorescence, rather than 1:1 extract analysis, provided a greater understanding of the overall contamination source presented by the solid tailings. X-ray fluorescence data for uncontacted tailings, leached tailings, and laboratory prepared clay liner are shown in Table 22.

Results of X-ray fluorescence analysis showed As, Cl, Cr, Cu, Mo, Ni, Pb, U, V, and Zn were present in the uncontacted tailings and leached tailing at concentrations several orders of magnitude higher than the uncontacted liner. With the exception of As, these constituents indicated reduced concentrations in the leached tailings in comparison with the uncontacted tailings, suggesting they may have been removed from the contacted tailings as part of the leachate front.

TABLE 22. X-Ray Fluorescence Data Indicating Concentrations of Solid Material for Uncontacted and Leached Tailings and Laboratory Clay Liner

<u>Parameter</u>	<u>Uncontacted Tailings, ppm</u>	<u>Leached Tailings, ppm</u>	<u>Laboratory Clay Liner, ppm</u>
Al	49,800	47,300	40,090
As	113.9	373	7.21
Br	<0.91	<1.1	2.12
Ca	36,900	27,600	90,900
Cl	2,640	640	610
Co	27	24	25
Cr	105	66	34
Cu	56	21.9	13.9
Fe	20,900	18,380	16,300
K	8,620	13,250	11,030
Mn	49.9	115	184
Mo	10.4	8.5	4.4
Ni	253	133	16.6
P	<0.33	<0.33	<0.33
Pb	201	76	14
S	26,100	13,220	44,500
Si	285,000	300,000	217,000
Sr	323	168	397
Ti	2,300	2,100	1,780
U	201	57.5	4.9
V	1,683	1,101	36
Zn	109.5	87.2	63.3
Zr	201	174	165

On the other hand, concentrations of Ca, K, Fe, Mn, and S (most likely in the form of sulfates) are conspicuously high in the uncontacted clay liner. The elevated sulfate and iron concentrations in the uncontacted clay liner could mask any movement of these constituents through the liner system. The two-fold drop in sulfate concentration in the leached tailings extracts in comparison with the uncontacted tailings suggests sulfate movement out of the tailings. X-ray fluorescence data indicating concentrations of solid materials for Core No. 5 subsample are shown in Table 23.

The X-ray fluorescence data for Core No. 5 subsamples showed elevated concentrations of As, Cl, Cr, Cu, Ni, Pb, U, and V above the liner. Of these species only Cl and Pb maintained elevated concentrations below the clay liner. In particular, As, Ni, U, and V either remained insoluble or were well attenuated by the clay liner. Again, levels of Ca, K, and Mn were higher in the clay liner and foundation soil than in the solid tailings.

Chemical analyses of the 1:1 extracts produced for uncontacted tailings, leached tailings, laboratory-produced clay liner and uncontacted overburden are given in Table 24, which includes the results of radionuclide counting on solid samples and evaporite salts of each tailings sample. Analytical results were similar to those obtained by X-ray fluorescence. Constituents As, Cl, K, Mg, Mo, Sr, Si, V, and Zn were present in solution at elevated concentrations from the tailings extracts in comparison with the uncontacted liner and foundation soil extracts. In addition, Cl, K, Mg, Mo, Na, Ni, SO_4 , Sr, V, and Zn appeared in higher concentration in the uncontacted tailings extract than the extract produced from leached tailings, suggesting that these species may be mobile in the system.

With the exception of ^{226}Ra , radionuclide counting showed that although measurable levels and radionuclide contamination were present in the solid samples, their solubility was very limited at the neutral pH of the liner system. The ^{226}Ra present in the extract of the uncontacted tailings extract seemed rather anomalous, considering that the levels in the leached tailings extract were below detection limits and both samples had equal concentrations in the solid samples.

TABLE 23. X-Ray Fluorescence Data for Core No. 5 Subsamples

Parameter (ppm)	Column Top to 24.38 cm	24.38 cm to 18.29 cm	18.29 cm to 12.19 cm	12.19 cm to 6.10 cm	6.10 cm to Column Bottom
Al	36100	50700	50000	50300	56000
As	76.1	62.6	13.5	8.5	8.9
Br	<1.0	<1.0	1.24	1.22	<1.0
Ca	25,500	25,900	33,200	31,900	56,700
Cl	620	680	720	640	620
Co	<22	<24	<26	<27	<28
Cr	51	60	54	36	43
Cu	22.1	19.8	20.6	19.7	18.0
Fe	15,140	17,560	20,100	22,200	22,400
K	12,510	17,220	16,960	16,090	15,180
Mn	109	155	188	210	223
Mo	9.1	6.3	7.4	5.9	5.5
Ni	58.5	72.2	21.6	21.4	18.2
P	3000	3100	3100	3100	3300
Pb	62.4	58.1	39.2	30.4	25.9
S	9740	9070	7580	8620	7350
Si	322,000	308,000	287,000	281,000	270,000
Sr	144	156	146	155	199
Ti	1770	1990	2290	2220	2270
U	44.1	32.5	6.7	5.3	5.0
V	785	565	79	48	47
Zn	97.6	95.4	82.3	83.7	79.1
Zr	202	210	218	223	220

TABLE 24. Chemical Analysis of Tailings and Uncontacted Liner Material:
1:1 Extracts

	Uncontacted Tailings	Leached Tailings	Laboratory Prepared Clay Liner	Uncontacted Foundation Soil
Macro ions (mg/L)				
Al	0.11	0.05	0.08	0.06
B	0.43	0.43	0.37	0.04
Ca	680	804	470	562
Fe	0.11	0.07	0.04	0.12
K	71.0	37.1	13.1	7.4
Mg	517.75	89.4	19.6	26.0
Na	1,677	259.3	1370	1610
Si	39.6	33.0	9.3	5.1
Sr	8.80	4.91	5.7	4.6
Cl	2400	170	130	450
NO ₃	1100	ND(a)	770	1100
SO ₄	3,200	2,500	4,400	4850
Trace metals (mg/L)				
As	1.74	1.56	0.06	0.1
Ba	0.06	0.04	0.03	0.04
Cr	0.08	0.06	0.02	0.02
Mo	3.60	1.05	0.16	0.03
Ni	0.29	0.00	0.06	0.03
Se	3.14	3.58	0.50	0.00
V	8.73	6.58	0.01	0.01
Zn	0.46	0.22	0.08	0.08
Radionuclides solid samples (pCi/g)				
230Th	467	500	ND	ND
210Pb	511	509	ND	ND
226Ra	734	710	ND	ND
235U	<50	<50	ND	ND
Radionuclides 1:1 Extracts (pCi/L)				
230Th	<50	<50	ND	ND
210Pb	<50	<50	ND	ND
226Ra	419	<50	ND	ND
235U	<50	<50	ND	ND

(a) ND = Not Determined

Extracts produced from each core subsample were chemically analyzed, and the averaged results for each depth are reported in Table 25. Radionuclide data were obtained by counting solid subsamples from Core No. 5. Initial counting data for 1:1 extracts produced from Core No. 5 subsamples showed no measurable radionuclides in solution at any depth.

Results of the chemical analysis of the 1:1 extracts from the core subsamples indicated movement of Cl and NO_3 through the entire liner system. These results were expected because the solubilities of Cl and NO_3 are not limited in neutral pH conditions. Concentrations of K, Mg, and Sr were elevated in and below the clay liner, indicating that they were passing through the liner with little attenuation. While Ca, Na, and SO_4 concentrations were high throughout the liner system, the source of these contaminants was in doubt due to their elevated concentrations in both the liner mix and the foundation soil. The clay liner did, in fact, attenuate As, Mo, Si, Se, and V because clear reductions in concentrations can be seen below the clay liner depth. This was consistent with the leachate analysis collected from the moisture extraction cups and piezometers during the field tests. Radionuclide results also showed clear concentration reductions below the clay liner for all species measured.

The large variations in the reported results for extract analysis reflected the fact that the major form of attenuation provided by the clay liner in a neutral environment was adsorption by clay minerals. This form of attenuation has a finite ability to entrap contamination and its capabilities are enhanced by low leachate flow rates. Thus, variations in reported concentrations were most likely due to variations in the permeability of the liner system from location to location as shown in the core permeability measurements.

CONCLUSIONS ON FIELD LINER EVALUATIONS

Analysis of the moisture block and moisture extraction cup data thus far shows that the asphalt membrane and clay liner materials provided an effective contaminant barrier at the Grand Junction test pit.

TABLE 25. Chemical Analysis of Core Subsamples: 1:1 Extract

	Column Top to 24.38 cm	24.38 cm to 18.29 cm	18.29 cm to 12.19 cm	12.19 cm to 6.10 cm	6.10 cm to Bottom
Macro ions (mg/L)					
Al	0.05 ±0.06	0.24 ±0.37	0.02 ±0.03	0.02 ±0.03	0.00
B	0.42 ±0.19	0.27 ±0.21	0.49 ±0.12	0.45 ±0.007	0.46 ±0.02
Ca	1029.3 ±110.03	910.7 ±196.7	1375.5 ±472.05	1286.0 ±441.27	1282.75 ±453.64
Fe	0.05 ±0.02	0.06 ±0.004	0.05 ±0.01	0.11 ±0.12	0.06 ±0.03
K	64.80 ±8.13	72.91 ±2.91	103.08 ±9.68	111.66 ±31.23	121.5 ±47.4
Mg	98.8 ±25.9	151.3 ±44.1	230.2 ±20.5	242.1 ±69.5	251.6 ±23.6
Na	935.0 ±217.3	1108.5 ±367.3	1393.1 ±392.6	1089.3 ±282.9	1209.6 ±101.6
Si	24.7 ±2.0	18.1 ±7.0	10.62 ±1.0	7.9 ±1.7	6.40 ±2.45
Sr	5.2 ±0.7	5.5 ±1.2	7.0 ±1.3	6.2 ±1.2	6.6 ±1.0
Cl	437 ±188	551 ±384	497 ±327	500 ±217	523 ±179
NO ₃	350 ±184	475 ±285	506 ±310	713 ±412	896 ±525
SO ₄	3367 ±153	4383 ±1487	4700 ±1345	5333 ±757	5300 ±819
Trace Metals (mg/L)					
As	0.55 ±0.42	0.24 ±0.20	0.06 ±0.03	0.05 ±0.05	0.08 ±0.06
Ba	0.04 ±0.01	0.04 ±0.01	0.06 ±0.01	0.05 ±0.01	0.03 ±0.03
Cr	0.03 ±0.01	0.02 ±0.02	0.04 ±0.02	0.04 ±0.02	0.04 ±0.02
Mo	1.53 ±0.03	1.28 ±0.71	0.74 ±0.48	0.59 ±0.38	0.16 ±0.13
Ni	0.06 ±0.01	0.03 ±0.01	0.02 ±0.04	0.03 ±0.01	0.03 ±0.01
Se	2.17 ±0.21	1.34 ±0.33	0.44 ±0.48	1.03 ±0.58	0.66 ±1.14
V	2.41 ±0.12	0.91 ±0.89	0.02 ±0.01	0.05 ±0.02	0.04 ±0.02
Zn	0.06 ±0.05	0.04 ±0.04	0.16 ±0.21	0.04 ±0.02	0.03 ±0.01
Radionuclides (pCi/g)					
²³⁰ Th	234	262	<50	<50	<50
²¹⁰ Pb	278	246	<50	<50	<50
²²⁶ Ra	361	359	<50	<50	<50
²³⁵ U	<50	<50	<50	<50	<50

The moisture block data indicates that the asphalt membrane effectiveness was due to its impermeable characteristics. Inspection of asphalt samples showed them to be lively, of adequate thickness, with small crust formation on the exposed surface and very few surface cracks. Although a small zone of high moisture foundation soil was observed, it was attributed to faulty installation and inspection practices. A list of required construction practices is provided in this report.

The test clay liner, while allowing leachate to permeate through it, was effective in reducing concentrations of most cation and trace metal contaminants through adsorption to clay minerals. Anions in solution, including Cl and NO₃, experience only limited attenuation while passing through the clay liner. These constituents will require specific treatment to ensure that their concentrations are below water quality guidelines. Radionuclide concentrations below the clay liner showed that no radionuclide movement away from the tailings pile was detected. It is probable that a more sensitive detection technique such as pulsed laser fluorimetry in neutral pH leachates would have confirmed that the clay liner was an effective radionuclide barrier.

The physical characteristics of the test clay liner indicated that it is possible to achieve a quality mixture of sodium bentonite and local soil in the field. Installation of the clay liner mixture under field conditions produced bulk density results ranging from 88 to 100% proctor for the samples tested. Because the hydraulic conductivity of the clay liner depends heavily on the bulk densities achieved in the field, the most effective method for improving contaminant migration through a sodium bentonite liner is to ensure that maximum compaction is achieved uniformly over the entire area.

DECISION TREE FOR LINER EVALUATION

The decision tree (Figure 37) 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.

SITE CHARACTERIZATIONS

If the preliminary site assessment identifies a possible groundwater problem, decision tree methodology can be used. The first step in the procedure is site characterization. Because of the initial expense in using liners or having to take remedial action, comprehensive site characterization should be completed prior to construction activities so that the decision tree can be used to determine whether site-specific objectives can be met. This will save time and money if the design is inadequate 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 exist 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 necessary data will be obtained.

The decision on liner emplacement will ultimately be based on predictions made with state-of-the-art computer models; however, they can only be 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. Site characterization data can be divided into three categories, which are discussed below:

1. site hydrology
2. tailings characterization
3. contaminant attenuation.

FIGURE 37. 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 Hydrology

Characterization of the site hydrology can be considered adequate if sufficient data exist 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.

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

Figure 38 shows the water balance for surface water at a hypothetical site. Precipitation and irrigation provide water to the surface, while runoff and evapotranspiration remove surface water. The remaining water 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 needed.

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 needed to calculate evapotranspiration. Such data include daily temperature fluctuations, wind speed, cloud cover, and thunderstorm events (amount of rain, time of day, and duration).

(a) These data can be obtained from:
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 38. Water Balance for a Hypothetical Uranium Mill Tailings Disposal Site (Below Grade Example)

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 required 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 is 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 can 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 that 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 (and shorter run time). Flux calculations from UNSAT1D for an average or maximum rainfall year are input to a model that describes water transport through the tailings and subsurface layers. One model with this capability is TRUST (Reisenauer et al. 1982).

TRUST can 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 boundary conditions needed for this and other suitable models are similar.

The following data are required for unsaturated subsurface hydrologic models:

- 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 ensure 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 as well because the subsurface and liner material samples needed for hydrologic characterization are also required to characterize leachate interactions with these materials.

One important interaction that is required 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 decrease as leachate passes through the column. Also, characterization of the inactive tailings may indicate that not all tailings require excavation and burial. Therefore, the next parameters are those that must be investigated to characterize tailings for the decision tree.

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 the history of 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, will help in predicting 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 Buel 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 is analyzed. Solid-to-solution ratios ranged from 1:5 to 1:100 (g tailings/cm³ water).

The batch method is acceptable qualitatively for screening a large number of tailings samples for relative contaminant concentrations, but it does not accurately determine how much contamination will be released from tailings and yields little (if any) information about contaminant release rates. Batch leach tests generally underestimate initial contaminant concentrations that may be leached from tailings upon contact with water.

The use of large columns (2000 cm³) allows analysis of radioactive and nonradioactive contaminants in the same sample with the precision needed to critically assess the first tailings leachate pore volume. Results indicate that effluent (leachate) from the tailings leach column should be

collected in three 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. The tailings characterization study showed that the contaminant concentrations fall rapidly within one or two pore volumes.

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, it must be assumed that contaminants migrate unattenuated with the same velocity as water through subsurface layers. This assumption is clearly in error and can lead to the construction of expensive and potentially unnecessary engineered barriers. To make realistic predictions of attenuated contaminant migration, the interactions of tailings leachate with soil and liner material must also be quantified.

Contaminant Attenuation

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, and this common link between solute (contaminant) and water transport is the retardation factor, which is discussed 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 nonlinear adsorption in saturated and unsaturated porous media is given by Persaud and Wierenga (1982).

Neither of these two transport models (or any other model) is presently able to predict solute/contaminant transport adequately 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, Reisnauer, and Gee 1980). To predict contaminant migration, two retardation factors are required for each contaminant: one retardation factor that describes 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 contacts 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 can be expressed by

$$R_f = 1 + \frac{\rho^b}{\theta} R_d \quad (7)$$

where 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). The approach of Nelson et al. (1980) will be used here to define an R_d value for the special case of pH.

The titration curve, shown in Figure 23, illustrates the change in the pH of a Clive, Utah, soil as leachate from Shiprock tailings was added. This study and those of Gee et al. (1980) have shown that many of the contaminant concentrations are greatly reduced at pH 7.7; therefore, R_d will be calculated for the pH at pH = 7.7.

Acidity (high H^+ concentrations) may be thought of as a contaminant because highly acidic conditions increase 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 per liter. In addition to H^+ , the total acidity includes Al^{+3} and Fe^{+3} because at higher pH values these hydrous oxides are precipitated and release hydrogen ions equivalent to the original Al and Fe concentrations. From Table 13 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 grams (see Figure 33). The resulting R_d for Shiprock leachate at pH = 7.7 in this soil is

$$R_d = \frac{230 \text{ meq/100 g}}{300 \text{ meq/1000 cm}^3} = 7.7 \text{ cm}^3/\text{g}$$

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

If pH = 4.5 had been selected for the above example, the result would have shown that 100 grams of soil neutralizes 470 meq of acidity. The R_d value for pH = 4.5 is

$$R_d = \frac{470 \text{ meq/100 g}}{300 \text{ meq/1000 cm}^3} = 15.7 \text{ cm}^3/\text{g} .$$

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 (7). If the soil bulk density is 1.5 mg/cm^3 and porosity is 0.4, the pH 7.7 retardation factor is

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

or

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

In other words, the Shiprock leachate pH front (at pH 7.7) moves 30 times slower than 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, two R_d values are needed for each contaminant. For this example, one R_d 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 R_d values.

A known volume of leachate (V) was added to a known amount of soil (W), and contaminant concentrations were measured before contact (C_0) and afterward (C_f). The amount of contaminant adsorbed on or precipitated in

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

$$R_d = \frac{(C_o V - C_f)/W}{C_f} \quad (8)$$

or more simply,

$$R_d = \frac{V}{W} \left(\frac{C_o}{C_f} - 1 \right) .$$

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 proceed to the second decision point.

WATER MOVEMENT WITHOUT A LINER

This step requires an experienced groundwater 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. (1980a), 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, 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, contaminant transport must be predicted.

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, no liner is required for tailings disposal at that site. If the answer is no, contaminant levels above the limits are predicted to cross the boundary for an unlined tailings pile; thus, either 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.

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, and a new computer simulation of water movement with the liner must be run and compared with the design objectives. The question now is: 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,

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, 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, Buelt, and Hale (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.

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 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, 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.

DEMONSTRATION

Relyea (1983) has demonstrated the decision tree for a hypothetical disposal site. The demonstration also showed that infiltration through the

cover is difficult unless composed of porous sand or rock that reduces evapotranspiration. Therefore, infiltration at Western U.S. sites may not be likely and the only water source may be from movement of a shallow groundwater table. Nevertheless, the demonstration, using the UNSAT1D and TRUST codes, compared natural soil and liners as contaminant retardation barriers for a variety of configurations and leachates. It showed that the decision tree is a logical sequence that can be helpful in determining liner requirements for a mill tailings impoundment.

cover is difficult unless covered by sand or rock that reduces
water infiltration. Therefore, infiltration at water table may not
be likely and the only water source may be from movement of a shallow
groundwater table. Nevertheless, the degree of infiltration using the UNSAT-1D and
US2 codes compared natural soil and there is continuous restoration
process for a variety of conditions and features. It showed that the
infiltration rate is a function of degree that can be helpful in determining
their requirements for a high quality environment.

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APPENDIX A

PERMEABILITY TEST DATA

TABLE A.1. Laboratory Permeability Testing Data

Test Number	Asphalt Cement					Asphalt Rubber				
	ΔV , L	Δt , h	P, psig	Th, in.	k (cm/s)	ΔV , L	Δt , h	P, psig	Th, in.	k (cm/s)
1 (4/23)	2	191	15	3	7.20×10^{-9}	19.8	23.63	15	0.28	5.31×10^{-8}
2 (5/7)	1	116.75	10		8.84×10^{-9}	6	22.64	10		2.52×10^{-8}
3 (5/12)	--	--	--		--	35.5	21.92	10		1.53×10^{-7}
4 (5/19)	3.25	169.3	11.5		1.72×10^{-8}	11.76	5.58	11.5		1.74×10^{-7}
5 (5/27)	9.75	166.5	11.5		5.25×10^{-8}	10.22	0.5	11.5		1.69×10^{-6}
6 (6/3)	7.5	52.08	10.5		1.42×10^{-7}	14.84	1	10.5		1.34×10^{-6}
7 (6/10)	--	--	--		--	--	--	--		--
8 (6/11)	5.8	73.60	11		7.40×10^{-8}	15.12	0.33	11		4.36×10^{-6}
9 (6/16)	4.26	68.72	11		5.81×10^{-8}	17.06	0.4	11		3.68×10^{-6}
10 (6/26)	7.64	67.68	11		1.06×10^{-7}	16.8	0.42	11		3.45×10^{-6}
11 ((6/26)	6.10	95.5	11		5.99×10^{-8}	16.90	0.37	11		3.95×10^{-6}
	Catalytic Airblown					Hypalon				
	ΔV , L	Δt , h	P, psig	Th, in.	k (cm/s)	ΔV , L	Δt , h	P, psig	Th, in.	k (cm/s)
1 (4/23)	5.6	23.05	15	0.25	1.39×10^{-8}	0.5	191	15	0.065	3.9×10^{-11}
2 (5/7)	4.12	22.62	10		1.57×10^{-8}	~0	116.75	10		$<4.79 \times 10^{-11}$
3 (5/12)	1.1	22.42	10		4.22×10^{-9}	--	--	--		--
4 (5/19)	2.13	26	11.5		6.12×10^{-9}	1.5	169.3	11.5		1.72×10^{-10}
5 (5/27)	6.32	47.7	11.5		9.91×10^{-9}	1.25	166.5	11.5		1.46×10^{-10}
6 (6/3)	0.5	22.5	10.5		1.82×10^{-9}	3.5	147.63	10.5		5.05×10^{-10}
7 (6/10)	--	--	--		--	--	--	--		--
8 (6/11)	3.8	4.53	11		6.56×10^{-8}	0	73.60	11		$<6.9 \times 10^{-11}$
9 (6/16)	6.86	68.72	11		7.8×10^{-9}	1	169.1	11		1.20×10^{-10}
10 (6/23)	15	163.2	11		7.18×10^{-9}	--	--	--		--
11 (6/26)	--	--	--		--	1	94.62			2.15×10^{-10}

TABLE A.1. (Contd)

A.2

Test Number	Bentonite					Saline Seal 100				
	ΔV , L	Δt , h	P, psig	Th, in.	k (cm/s)	ΔV , L	Δt , h	P, psig	Th, in.	k (cm/s)
1 (4/23)	18.8	23.05	15	1	$1.87 (10^{-7})$	9.7	0.5	15	1	$9.08 (10^{-6})$
2 (5/7)	4.96	1.65	15		$6.89 (10^{-7})$	18.5	1	17		$3.74 (10^{-6})$
3 (5/12)	18.1	21.92	12		$2.37 (10^{-7})$	11.2	0.5	13.3		$5.79 (10^{-6})$
4 (5/19)	4.62	5.58	10		$2.85 (10^{-7})$	13.8	0.75	10		$6.33 (10^{-6})$
5 (5/27)	5.24	3.88	10.5		$4.42 (10^{-7})$	15.1	1	10.5		$4.94 (10^{-6})$
6 (6/3)	13.3	22.5	11		$1.85 (10^{-7})$	16.6	1	11		$4.71 (10^{-6})$
7 (6/10)	6.56	23	11		$8.92 (10^{-8})$	16.83	0.92	11		$5.72 (10^{-6})$
8 (6/11)	6.62	23.17	11		$8.93 (10^{-8})$	15.94	0.58	11		$8.59 (10^{-6})$
9 (6/16)	8.72	23	11		$1.19 (10^{-7})$	16.68	0.62	11		$8.46 (10^{-6})$
10 (6/23)	11.88	24	11		$1.55 (10^{-7})$	16.32	0.63	11		$8.10 (10^{-6})$
11 (6/26)	10.81	20.75	11		$1.63 (10^{-7})$	10.54	0.5	11		$6.59 (10^{-6})$
	GSR-60					Soil				
	ΔV , L	Δt , h	P, psig	Th, in.	k (cm/s)	ΔV , L	Δt , h	P, psig	Th, in.	k (cm/s)
1 (4/23)	19.3	2.42	15	1	$1.83 (10^{-6})$	15.8	0.5	15	1	$7.24 (10^{-6})$
2 (5/7)	15.26	1	17		$3.09 (10^{-6})$	14	0.37	17		$7.65 (10^{-6})$
3 (5/12)	7.7	1	12.5		$2.12 (10^{-6})$	14.5	0.42	10		$1.19 (10^{-5})$
4 (5/19)	14.75	2.75	10		$1.84 (10^{-6})$	12.0	0.5	10		$8.25 (10^{-6})$
5 (5/27)	16.22	2	10.5		$2.66 (10^{-6})$	16.18	0.5	10.5		$1.06 (10^{-5})$
6 (6/3)	15.8	1.5	11		$3.16 (10^{-6})$	18.25	0.5	11.0		$1.14 (10^{-5})$
7 (6/10)	16.86	1.5	11		$3.51 (10^{-6})$	--	--	--		--
8 (6/11)	16.44	0.66	11		$7.79 (10^{-6})$	--	--	--		--
9 (6/16)	16.24	0.78	11		$6.48 (10^{-6})$	--	--	--		--
10 (6/23)	16.42	0.9	11		$5.70 (10^{-6})$	16.12	0.42	11		$1.20 (10^{-5})$
11 (6/26)	7.5	0.5	11		$4.69 (10^{-6})$	16.64	0.47	11		$1.11 (10^{-5})$

APPENDIX B

ASPHALT LINER SPECIFICATIONS

ASPHALT LINER SPECIFICATIONS

TABLE B.1. AR4000 Asphalt Cement Specifications

<u>Property</u>	<u>Minimum</u>	<u>Maximum</u>	<u>Actual</u>
Tests on original asphalt:			
Flash point, P-M, °F	440		480
Solubility in trichloroethylene, %	99		99.9
Tests on residue from RTFC:			
Absolute viscosity @ 140°F, ps	2500	5000	3260
Kinematic viscosity @ 275°F, cs	275		414.6
Penetration @ 77°F, 100 g, 5 s (tenths of mm)	40		53
Ductility @ 45°F, (1 cm/min) cm	10		50+

TABLE B.2. Hydraulic Asphalt Concrete Aggregate Specifications

<u>Grid Size</u>	<u>% Passing</u>
1/2	100
3/8	100
4	77
8	60
10	54
30	44
50	23
100	17
200	15

TABLE B.3. Specifications of Catalytic Airblown Asphalt

Catalytic Airblown Asphalt
ASTM No. D2521

Property	Specification		Typical Analysis
	Minimum	Maximum	
Softening Point (°C) (175°F)	79 (175°F)	93	88
Penetration @25°C (77°F), 100 g, 5 s	50	60	56
Penetration @0°C (32°F), 200 g, 60 s (tenths of mm)	30		32
Penetration @46°C (115°F), 50 g, 5 s (tenths of mm)		120	114
Flash Point (°C)	218		232
Solubility in Trichloroethylene (wt%)	97.0		99.0
Ductility @25°C (77°F), 0.083 cm/s (cm)	3.5		6.0
Specific Gravity @25°C (77°F)			1.027
Specific Gravity @15.6°C (60°F)			1.031
Penetration @25°C (77°F), 100 g, 5 s after heating to 163°C for 5 hours (% of original)	60		72
Loss @163°C, 5 hours (wt%)		1.0	0.1

APPENDIX C

EXPOSURE CONDITIONS, PERMEABILITIES,
AND REACTION RATES

APPENDIX C

EXPOSURE CONDITIONS, PERMEABILITIES, AND REACTION RATES

This appendix presents the exposure conditions, permeabilities, and integrated infrared absorption band data for the aged liners. Reference Codes A, B, and D through G refer to the first set of exposure tests. Codes 1 through 7 refer to the second set of exposure tests; and Code Z is the control, unexposed asphalt.

FIGURE C.1. Exposure Conditions for Liner A

FIGURE C.2. Exposure Conditions for Liner B

FIGURE C.3. Exposure Conditions for Liner D

FIGURE C.4. Exposure Conditions for Liner E

FIGURE C.5. Exposure Conditions for Liner F

FIGURE C.6. Exposure Conditions for Liner G

FIGURE C.7. Exposure Conditions for Liner 1

FIGURE C.8. Exposure Conditions for Liner 2

FIGURE C.9. Exposure Conditions for Liner 3

FIGURE C.10. Exposure Conditions for Liner 4

FIGURE C.11. Exposure Conditions for Liner 5

FIGURE C.12. Exposure Conditions for Liner 6

FIGURE C.13. Exposure Conditions for Liner 7

FIGURE C.14. Permeabilities of Liners Exposed To Low Temperature Conditions

FIGURE C.15. Permeabilities of Liners Exposed to Intermediate Temperature Conditions

FIGURE C.16. Permeabilities of Liners Exposed to High Temperature Conditions

FIGURE C.17. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner A

FIGURE C.18. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner B

FIGURE C.19. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner D

FIGURE C.20. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner E

FIGURE C.21. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner F

FIGURE C.22. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner G

FIGURE C.23. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into the Control Liner Z

FIGURE C.24. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner 1

FIGURE C.25. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner 2

FIGURE C.26. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner 3

FIGURE C.27. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner 4

FIGURE C.28. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner 5

FIGURE C.29. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner 6

FIGURE C.30. Integrated Absorbance in the Carbonyl and Carbon Double Bond Regions as a Function of Depth into Liner 7

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