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A STUDY OF SHALLOW LAND BURIAL OF LOW-LEVEL RADIOACTIVE WASTE

Annual Report for
Fiscal Year 1979-80

Prepared for

LOS ALAMOS SCIENTIFIC LABORATORY

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Submitted By

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INTRODUCTION

1.1 Background and Purpose

Since February 1977, the Civil Engineering Department at the University of Texas at Austin has been engaged in a research project with the Los Alamos Scientific Laboratory (LASL) for the Department of Energy (DOE). The purpose of this project is to develop improved designs for containment of low-level radioactive wastes in shallow land burials. The designs should be both economical and effective in eliminating outmigration of pollutants for a reasonable number of half lives of the principal radionuclides. Although the direct interest is in low-level radioactive wastes, the research findings will be applicable to disposal of non-radioactive wastes, as well. In addition to developing new designs, or improving upon existing ones, we are also to assess the probably cost and effectiveness associated with the design.

The first phase of this project (February through September 1977) involved acquisition of background information from the literature, from contacts with other researchers involved with similar or allied studies, and from visits to burial sites and discussions with their personnel. From information obtained during Phase I, it was concluded that one of the most economical engineered barriers available for retarding migration of radionuclides out of disposal pits is compacted soil. Consequently, a substantial portion of the subsequent work on the project was focused on design of soil barriers, either with or without synthetic materials, or clays mixed with local soil or placed between layers of local soil.

Phase II of the project (October 1977 through September 1978) was concerned primarily with identificaiton of design parameters and, where necessary, development of apparatus to measure those parameters. Because of past problems with performance of sites in humid regions, both we and LASL agreed that it would be best for us to concentrate our efforts on sites located in relatively arid regions. Therefore, much of the Phase II work involved developing means for measuring the relevant hydraulic properties of unsaturated, compacted soil barriers of the type that might be used at LASL or some other arid site.

Phase III of the project (October 1978 to September 1979) was concerned primarily with development of preliminary design concepts and proof-testing of some experimental techniques for measurement of design parameters.

Phase IV of the project began in October 1979 and ended in September 1980. The purpose of this phase of work was as follows:

1. To continue developing designs for liners and caps for representative disposal sites, such as Los Alamos. The goal is to find a liner that will not crack when desiccated and will remain relatively impervious. Major emphasis was put on soil mixtures because they hold the most promise; however, synthetic materials were also considered.

2. To continue developing suitable methods for analyzing movement of fluids out of lined or unlined disposal pits.

3. To assist with a study of burying the wastes in deeper pits than are currently used.

4. To complete a study of pretreatment alternatives and a study of the effects of organic matter and biological activity on the mobilization radionuclides.

Results for Item 3 above are presented in a report being submitted separately. All other results are summarized in this report.

1.2 Publications

Three masters' theses were published during FY 1979-80:

1. "Biological Mobilization of Buried Low-Level Radioactive Wastes," by Miguel A. Herrera, Dec., 1979.

2. "Volume Reduction and Fixation of Non-High Level Radioactive Wastes," by David E. Sheppard, Dec., 1979.

3. "Evaluation of Caps and Liners for Low-Level Radioactive Waste Disposal Pits," by Martha Pertusa, Aug., 1980.

Two Geotechnical Engineering Reports were issued through our Department of Civil Engineering:

1. "Geotechnical Aspects in Design of Disposal Sites for Low-Level Radioactive Wastes," by D. E. Daniel and R. E. Olson, April, 1980, GR80-6.

2. "Materials to Line or to Cap Disposal Pits for Low-Level Radioactive Wastes," by Martha Pertusa, April, 1980, GR80-7.

Copies of these reports and theses accompany this annual report.

2.0

BARRIERS TO LINE OR CAP DISPOSAL PITS

Low-level radioactive wastes have traditionally been buried in unlined pits under a few feet of soil cover. At some sites, radioactive isotopes have been leached from the waste and have escaped from the pits in dilute concentrations. Offsite movement of radionuclides at several sites has resulted in the decision to close the site and to consider improved methods of disposal. One potential improvement in disposal techniques would be to provide caps to minimize inflow of water and liners to retard outflow of pollutants. Such retardation would presumably increase the on-site retention time to such an extent that almost all radioactive decay would occur prior to escape and would lead to such a slow rate of escape that adsorption and dilution would render the effluent harmless.

The following summarizes a study of barriers that might be used to line or cap these disposal pits (Pertusa, 1980). The study included literature and personal communication, but not experimental work, and was concerned heavily with existing knowledge and experience. Possible designs which might solve some of the current problems at low-level disposal installations are suggested.

2.1 Types of Barriers Considered

Three types of barriers were considered in this study:

(1) "synthetic membranes" -- typically rubber or plastic sheets no thicker than about 50 mils (1 mil = 0.001 inch);

(2) "rigid barriers" -- typically asphaltic or Portland cement concrete, or soil treated with asphalt, lime, or Portland cement;

(3) "compacted soil barriers" -- typically compacted local clay but perhaps local soil to which bentonite or other material has been added to reduce hydraulic conductivity.

2.2 Sources and Quality of Information

An attempt was made to locate information from manufacturers, individual researchers, and the literature. The manufacturers were generally able and willing to provide quantitative information on the mechanical properties of their product, and qualitative information on chemical resistance. They were generally unable or unwilling to provide quantitative data on chemical resistance or data on actual field performance. Individual researchers often proved to be suspicious and uncooperative, claiming that their data were proprietary or not suitable for distribution. Only a few studies have been reported in the literature. In particular, the reference by Kays (1977) was a source of practical information. However, in general, the data available on synthetic membranes are meager in quantity, especially data from field observations, for fluids other than water, and for times greater than a few weeks or months. The causes of the deficiency in information include the facts that synthetic liners have been used for only about a decade for sealing landfills, and that the suppliers are in competition with each other and wish to minimize releases of information that might aid competitors.

2.3 Synthetic Membranes

Synthetic membranes are flexible sheets of plastic, rubber, or combinations thereof. They have been used to line sanitary landfills, uranium mill tailings ponds, and ponds containing hazardous industrial waste.

Synthetic membranes that are classified as rubbers include:

- Butyl
- EPDM (ethylene propylene diene monomer)
- Neoprene

Those classified as plastics are:

- PE (polyethylene)
- PVC (polyvinyl chloride)
- CPE (chlorinated polyethylene).

Chlorosulfonated polyethylene (commonly called Hypalon®) is a cross between the plastic and elastomer materials. Another type of membrane, 3110, falls into the category of elasticized polyolefins.

Polymeric membranes can be fabricated with a sheet of reinforcing fabric, which is called "scrim", sandwiched between layers of the polymeric membrane. Some advantages of using reinforcing fabric include:

- Better dimensional stability;
- Increased tear and puncture resistance;
- Increased tensile strength; and
- Easier handling in the field.

Disadvantages include:

- Less elongation prior to rupture;
- Less conformity to ground irregularities;
- Less flexibility; and
- Greater cost.

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Nylon reinforcing is the most popular type; however, it has poor resistance to acidic solutions and to sunlight. Polyester resists acids and sunlight but is not as strong as nylon.

2.3.1 Field Performance

The main criteria for selection of a membrane are its service life in the field and its cost. Service life is difficult to evaluate because field observations do not extend beyond 10 to 15 years for sanitary landfills or uranium mill tailings ponds. Thus, the following comments relate mainly to short-term problems.

Premature failure of membranes in the field have been caused primarily by the following problems:

- Improper preparation of the site, installation of the membrane, or construction of seams;
- Degradation of the membrane due to environmental factors;
- Chemical incompatibility between waste and liner;
- Excessive settlement of the subgrade; and
- Effects of radiation.

2.3.2 Comparison of Membranes

Although polymeric membranes may provide temporary containment of the radioactive waste, it is doubtful that absolute zero leakage is attainable. For zero leakage the following sequence of events would have to occur. Not only must the membrane by produced without defect, i.e., no pinholes, constant thickness, etc., but also no mishaps can occur in the packaging or shipping processes. Field installation must occur without mechanical damage, adhesive problems, weather difficulties, subgrade imperfections, etc. Settlement must also occur as predicted. Based on

this preliminary study evaluating possible synthetic membranes to line the trenches at a low-level radioactive waste disposal site, the following are considered unacceptable candidates:

Butyl -- Quality field seams are difficult to acheive. It has poor resistance when exposed to even low-level radiation.

Neoprene -- The literature describing Neoprene appears to be contradictory. Its predicted life is too short to justify the initial cost.

PE -- Conventional PE is susceptible to mechanical damage during installation.

PVC -- There are a series of problems associated with loss of plasticizer.

There are no liners that are free of problems. However, pending further research, the following liners show potential to serve as barriers:

Hypalon® -- Although this liner has few problems associated with field installation and appears resistant to a variety of environmental factors, the properties of the membrane change under exposure to different chemicals.

High Density PE -- Although more research is needed, these liners may serve as possible barriers. They are rugged and the extrusion-welded seams seem to be of high quality. A wide variety of chemicals can be contained. Low amounts of radiation exposure also appear to have no degrading effect for PE materials, in general.

Elasticized Polyolefin (3110) -- There is little experience associated with this membrane (it was invented in 1975). However, its characteristics in terms of ease of installation and resistance to environmental factors appear promising.

At present, it appears the relatively short service life (approximately twenty years) of synthetic liners is one of the biggest obstacles to be overcome before polymeric liners can be considered as potential barriers at sites used for disposal of low-level radioactive wastes. Research must be performed to indicate how the properties of synthetic membranes change when exposed to various chemicals in the waste over a period of many years.

2.4 Asphalt

"Asphaltic concrete" is the proper name for a mixture of inert mineral matter and asphalt cement. The mineral matter is typically sand and/or gravel and is used to provide bulk and frictional strength. The asphalt is used as a binder (cement), thus giving cohesive strength, and to fill the voids between the mineral grains, thus reducing permeability. The term "asphalt emulsion" indicates a colloidal suspension of asphalt in water.

Paving asphaltic concrete is typically formed by mixing heated aggregate and asphalt and spreading the mixture in layers approximately three times the size of the coarsest aggregate. A layer is genearlly not less than 1-1/2 inches thick, and if it is more than three inches thick, it is usually placed in two layers in such a manner that the joints are staggered. The layers should be compacted using smooth steel-wheeled rollers.

Another form of asphaltic liner is hot-sprayed asphalt, which may be sprayed directly onto the soil or onto woven synthetic fiber (such as polypropylene), a jute mat, or woven or nonwoven glass fiber. Spraying the asphalt on a supporting fabric adds toughness and dimensional stability. Difficulties associated with the installation of sprayed membranes include not only the requirement of a large amount of special equipment but also the problems associated with achieving a uniform membrane. Bubbles, which

often form during application of hot sprayed asphalt, may later rupture, causing failure of the membrane. As a result, a minimum of three or four passes should be made (1-1/2 to 2 gal/yd^2) to attain a final thickness of 1/4 to 3/4-inch.

"Hydraulic asphalt" has a permeability that is one or two orders of magnitude less than that of paving asphalt. This reduction in permeability is achieved by controlling the gradation of the aggregate and asphalt content to obtain a nearly voidless structure after compaction. This type of asphalt is generally more difficult to handle in the field than paving asphalt.

Newly developed asphalt liners, which have more flexibility at low temperatures than regular paving asphalt, include catalytically-blown asphalt, which is produced by air blowing in the presence of a catalyst and asphalt-rubber, which consists of granulated rubber mixed with asphalt. It is thought that the extent cracking occurs in asphalt-rubber is less than that of regular asphalt because the rubber particles tend to minimize the propagation of cracking (Morris and McDonald, 1976).

2.4.1 Field Performance

Asphaltic liners work well only if they are properly constructed; careful attention to quality control is essential. Potential construction problems include:

- Wrong temperature of mix;
- Set-up after placement but before compaction;
- Improper compaction; and
- Poor joints.

After the membrane is installed, environmental factors may degrade it. Exposed asphalt does not age well in desert areas. Although unaffected by ozone, it will decay if exposed to sunlight, apparently as a result of loss of the more volatile fractions. The loss of components is accompanied by shrinkage and embrittlement of the material. A soil cover consisting of from 1 to 3 feet of graded sand and gravel will delay oxidation of the asphalt. However, the cover should not consist of fine-grained soils, which tend to shrink when dried, because they may cause tensile stresses on the surface of the membrane (Asphalt Institute, 1976).

2.4.2 Discussion

Asphalt membranes appear to have potential application as barriers for low-level radioactive wastes. Proof of the durability of asphalt can be seen from structures remaining centuries after they were built. For example, about 1300 B.C., Mesopotamians built a levee along the banks of the Tigris River at Assur with limestone blocks mortared with asphalt mastic and faced with bricks bonded and water proofed with asphalt mastic. Portions of the hydraulic facility are still in existence (Asphalt Institute, 1976).

Another attractive feature of asphalt is that it is essentially impermeable, provided its not degraded by chemicals or subjected to significant deformations.

In terms of overall effectiveness, expected life and cost, asphalt is the most promising of the man-made materials. However, a soil covering is imperative.

2.5 Concrete

2.5.1 Definitions

The term "concrete" will be used here in its colloquial sense to mean Portland cement concrete. Mixtures of Portland cement, sand, and gravel

are typically termed "plain concrete." Addition of tensile reinforcement, typically wire mesh, deformed steel bars, and/or steel cables, leads to "reinforced concrete." "Pneumatically-applied concrete" (often called "gunite") is applied by mixing sand, Portland cement, and water in a "gun" and blowing the mixture against a surface to form a dense, strong layer, typically 3/4 to 1-1/2 inches thick.

2.5.2 Applications and Problems

A great deal of experience is associated with the use of concrete in water retention facilities. One of the principal advantages of using a concrete liner and cap at a disposal site would be the structural integrity of the barrier system which would eliminate most of the problems caused by settlement of poorly compacted waste.

If properly formulated and constructed, it is possible to have hydraulic conductivities as low as 10^{-10} cm/sec. However, attaining this order of permeability in the field is doubtful. Shrinkage cracks typically form during curing, and construction joints present leakage problems. Also, even good quality concrete has the tendency to degrade in its waterproofing qualities over long periods (Kays, 1977).

Concrete liners without reinforcement are particularly vulnerable to settlement of the subgrade. Degradation of the material may occur if the adjacent soil exhibits shrink/swell behavior (Kays, 1977). Also, concrete may deteriorate when subjected to freeze/thaw or wet/dry cycles. Gunite, which has the advantage that it can be sprayed on vertical uneven surfaces, also has problems with cracking during the curing process or due to differential settlement. The use of a wire mesh in the gunite may reduce these problems (Kays, 1977).

Concrete is susceptible to chemical attack under both acid and alkaline conditions, and if exposed to soils containing sulfates will gradually dissolve (SCS Engineers, 1977). Unlike asphalt, however, it is essentially unaffected by most organic compounds.

Although concrete has proved its usefulness at several low-level radioactive waste disposal sites, e.g., Los Alamos, Oak Ridge, Hanford, Savannah River, where it serves as containers to store plutonium and other transuranic waste (SCS Engineers, 1977), it is felt there are other less costly alternatives which might be just as effective in retaining low-level nontransuranic waste.

2.6 Soil Cement

"Soil cement" is a mixture of pulverized soil, cement, and water. The amount of cement added varies from 3% to 20%. Best results are obtained when the soil is sandy and well-graded with a maximum aggregate size of about 3/4-inch. Depending on the quantity of cement and soil type, the hydraulic conductivity may vary from 10 cm/sec to 1.5×10^{-8} cm/sec (SCS Engineers, 1977).

Soil cement is not considered an adequate liner or cap for low-level radioactive waste disposal sites because transverse shrinkage cracks occur during curing. Also, because of its brittle nature, soil cement could be subjected to only a limited amount of differential settlement. It could be used for erosion control, although there appears to be some contradictory evidence concerning its degradation when subjected to wet/dry or freeze/thaw cycles.

2.7 Soil Barriers

Soils may be used as barriers to retard flow or as drains to channel or divert the movement of fluids. Relatively impervious barriers can be obtained by compacting fine-grained soils (such as clay), mixtures of soils, or combinations of soil and chemical admixtures. The advantages of using compacted clay are that clay is relatively inexpensive and is resistant to chemical attack by leachate. Unless the leachate is strongly acidic, longevity of a clay barrier is seldom an issue. Problems with using compacted clay include possible cracking of the clay as a result of settlement or desiccation, and loss of integrity as a result of penetration by plant roots or burrowing animals.

It may be possible to design a tolerably impervious barrier by mixing a combination of soils so that the hydraulic conductivity and the formation of shrinkage cracks are reduced simultaneously. A mixture consisting primarily of sand but with enough clay to fill the voids among the sand grains would be expected to have an acceptable hydraulic conductivity and to experience small changes in volume when subjected to cycles of wetting and drying.

Cracks also develop in the cap when it is subjected to differential settlement. Settlement is caused by compression of the waste under load, movement of fines into adjacent cavities, and decomposition of the waste. Disruption of the integrity of the soil cap by settlement has occurred at several disposal sites including ORNL, Hanford, Savannah River, and LASL (Deichman, 1979; Hooker, 1979). There are several options available for reducing the magnitude of the post-construction settlement. For example, balers can reduce the volume of the waste by factors ranging from 2 to 10. A more economical solution might be preloading a recently finished trench with

a surcharge of soil. Another possibility is packing soil in the void spaces between containers of waste. A coarse-grained soil, e.g., sand or gravel, has the advantage of serving as a drain so that incoming moisture is in contact with the waste a minimum amount of time. As a result, the chances that the water will become contaminated are lessened.

Besides cracking, other problems include penetration of the compacted soil barrier by burrowing animals or by plant roots. The problem of penetration by burrowing animals can be reduced by noting that most subsurface organisms live within 3 to 4 feet of the ground surface. Penetration by animals could be expected to be negligible if the depth of burial is increased beyond the depth used in current practice.

However, plant root penetration still poses a problem because in arid regions, root systems may penetrate as much as 30 to 50 feet below the surface. Use of herbicides or layers of cobble or gravel buried at depth may be effective in deterring growth of plant roots (Cline, 1979).

In terms of chemical compatibility, most soils have good resistance to deterioration from chemical attack. Experiments described by Crim, Shepherd, and Nelson (1979), in which clays were permeated with acids having a pH of less than 1 for periods up to 9 months, indicate that clays can be remarkably stable in the short-term, even in exceedingly harsh chemical environments. However, long-term attack by strong acids (pH less than about 2) may result in a gradual breakdown of the structure of some soils (Moore, 1979).

Although more research is needed, compacted soil barriers appear to be a cost-effective solution for retarding the migration of radionuclides. Soils are stable materials which often act chemically to neutralize

escaping materials. Polyvalent cations are likely to be adsorbed by soil, and organics may be broken down by soil organisms. An increase in the thickness of the cap would reduce the possibilities of cracks penetrating to the waste or of plants or animals coming into contact with the refuse.

2.8 Drainage with Wick Concept

Because it is not possible with current technology to design a compacted soil cap that is 100% impervious for the length of time necessary to contain low-level radioactive waste, an alternative approach would be to design a cap that diverts infiltrating moisture away from the trench. One such approach termed the "wick effect" by Corey and Horton (1969) is based on the assumption that as long as the pore pressures along the wick-gravel interface are negative, water will not flow from an unsaturated finer-grained soil into an underlying gravel layer. As illustrated in Fig. 2.1, the essential features are a gravel layer surrounding the buried waste with a fine-grained soil overlying and surrounding the gravel. In their analysis, Corey and Horton made several simplifying assumptions, such as assuming the system is in a steady state. They concluded that an overlying gravel layer would not divert water around large burial trenches during periods of prolonged rain.

The wick effect was reexamined by Frind et al., (1979) using a finite element transient saturated-unsaturated flow model. Contrary to Corey and Horton's observations, Frind et al. concluded that the wick effect has potential usefulness in the storage of radioactive waste. They found that the extent to which water can be diverted while maintaining a negative pore pressure at the soil-gravel interface depends on the slope of the interface, the magnitude of the lateral hydraulic gradient, and the thickness of the wick, as well as the rainfall intensity and duration. In general, the

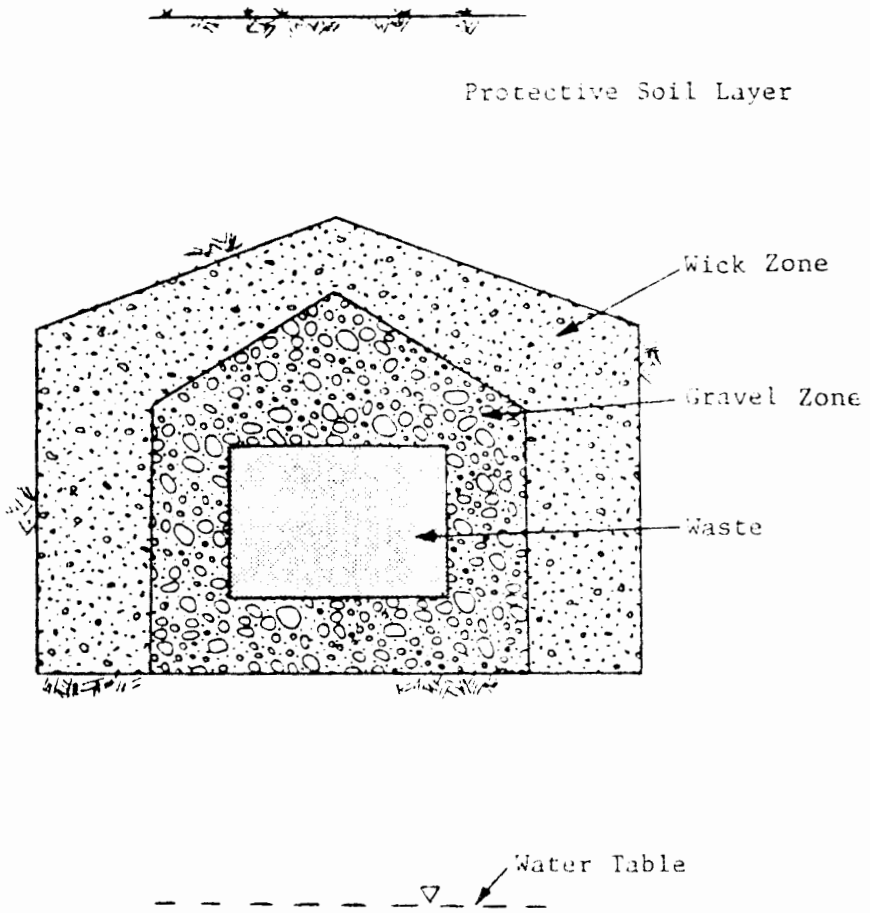


Fig. 2.1 Schematic drawing of the "wick effect" (Frind et al., 1979).

maximum pore pressure along the interface decreases as the slope of the interface and the thickness of the wick increase.

Application of the wick effect for the disposal of low-level radioactive waste should be at a site which has a relatively homogeneous soil of low permeability. The locale should have a dry climate with a deep water table which does not fluctuate significantly. Some field experimentation has been performed by Rancon (1979).

There are some possible problems associated with the construction and long-term integrity of such a facility. For example, the use of a filter may be necessary to maintain segregation of the coarse and fine-grained soils at the interface both during and after construction. There is also a question about the ability of a fine-grained soil to drain adequately so that it does not become saturated during periods of prolonged rain.

2.9 Comparison of Possible Barriers and Recommendations

2.9.1 Comparison of Materials

Important criteria in selecting a material to line and cap a disposal pit include the expected service life and the associated costs. Such a comparison is presented in Fig. 2.2. Although many of these materials have been used to line sanitary landfills, and ponds containing hazardous industrial waste or uranium mill tailings, by far the largest amount of experience has been associated with the use of these materials in water retention facilities. The life expectancies which have been conservatively estimated for water retention structures may be reduced by variable amounts if the material is exposed to acidic effluent, salts, organic solvents, etc., or if the material is subjected to significant amounts of differential settlement.

Fig. 2.2 Comparison of expected service life versus In-place cost (1976). (Service life estimate is for water retaining structures; + no estimate is available because membrane was invented in 1975. Unless noted, the cost estimate includes excavation, installation, backfilling, compaction, and seeding (by Dames and Moore, 1976); # cost adjustment made to include construction of subgrade and earth cover.)

Past experience indicates that synthetic membranes, in general, have an expected life of around twenty-five years. Using polymeric membranes to contain wastes is a relatively new field with little published information indicating how the membranes will withstand the environmental or chemical stresses that they may be subjected to over a span of years. Although some research is underway to determine the long-term effects of different chemicals and leachate on the membranes, these experiments have lasted, at most, three years which is far too short a time to provide data of the type needed. Although these membranes may provide temporary containment of radioactive waste, they do not appear to provide optimum containment for their cost. At present, synthetic membranes seem to have their main application for storage ponds, where location and repair of leaks is simplified.

Much more experience is associated with the use of concrete and asphalt not only with hydraulic facilities but also in radioactive waste disposal. For example, pretreatment of liquid radioactive waste may involve mixing it with polymer impregnated concrete or bitumen to form a matrix. Asphalt, however, appears to be more cost-effective in retaining radioactive waste than concrete. As illustrated in Fig. 2.2, there is a fairly significant variation in price depending on the type of asphalt, with paving asphalt being the most economical, and catalytically-blown asphalt the most expensive. The life expectancy of exposed asphalt is only on the order of 20 years. However, its estimated service life can be greatly increased with the use of proper soil covering and construction techniques. One disadvantage associated with the use of asphalt as opposed to concrete is the possible degradation of asphalt by organic compounds in the waste.

At present, soil barriers appear to offer the most economical solution for a given containment time of the waste. Unlike man-made materials which

deteriorate with time, soils are extremely stable. Evidence of the stability and attenuation capabilities of soils is demonstrated by the "Oklo phenomena." Around 2 billion years ago, a naturally occurring geological reactor began to produce fission products in what is now Oklo, Gabon (Africa). During this time span, as much as 10 metric tons of fission products were successfully stored in the ground. Most of the heavy elements remained relatively fixed. Although some radionuclides including Kr, Xe, Rb, Cs, Sr, Ba, Mo, and I have migrated during this period (Walton and Cowan, 1975), soil was generally very effective in retarding movement of radionuclides.

2.9.2 Recommendations

In regions with a fairly deep water table, soil barriers and drains may be incorporated into the design of caps so that the amount of moisture reaching the waste is significantly reduced. If moisture could be prevented from reaching the waste, the formation of leachate and subsequent migration of radionuclides would be minimal.

Caps. Design features should include surface drains which minimize the amount of moisture that penetrates into the ground. Once the moisture has infiltrated into the soil, subsurface drains should be used to divert the water away from the disposal pit. Any moisture that does reach the disposal pit should be channeled into internal drains within the pit so that incoming moisture is in contact with the waste a minimum amount of time. As a result, the chances that the moisture will be contaminated are lessened. Such a design is illustrated in Fig. 2.3.

For unsaturated conditions, any infiltrating moisture would be diverted laterally in the soil overlying the gravel layer, i.e., the flow would be governed by the wick effect. With saturated conditions, water would flow

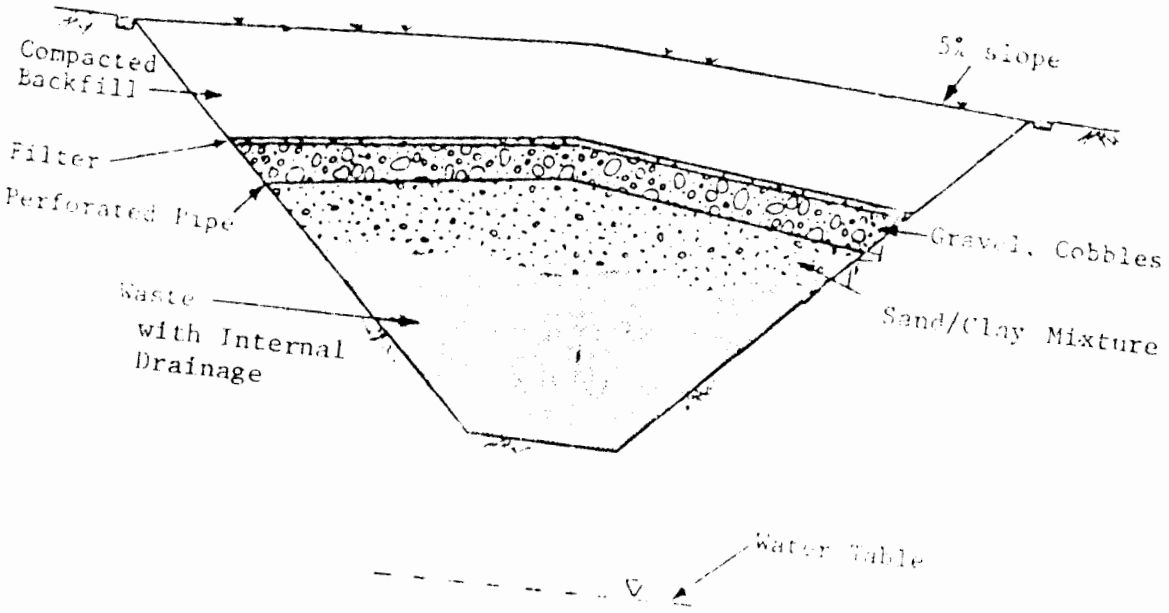


Fig. 2.3 Compacted soil cap with drain

into the gravel layer which would serve as a drain channeling the water away from the disposal pit. Other advantages of the layer of cobbles or gravel include the minimization of plant root penetration (Cline, 1979), and the deterrence of burrowing animals (Brunner and Keller, 1972; Cline, 1979). The gravel layer might also serve as a collection system for escaping radioactive gas. Use of vents and appropriate filters would allow the gas to be collected and treated.

In order for the gravel layer to fulfill its design functions it must be maintained free of fines. A graded filter thick enough to remain effective if post-construction settlement occurs would be necessary to prevent the overlying soil from migrating into the gravel.

The sand-clay layer below the layer of gravel (Fig. 2.3) would serve as a tolerably impermeable barrier that would guide infiltrating moisture away from the trench during prolonged rains. As mentioned previously, a sand-clay mixture would be expected to undergo little cracking when exposed to cycles of wetting and drying. Problems could arise if cracks occur in the sand-clay layer and coarse-grained soil migrates into the cracks forming permanent porous channels. Use of a filter cloth would eliminate this problem.

Liners

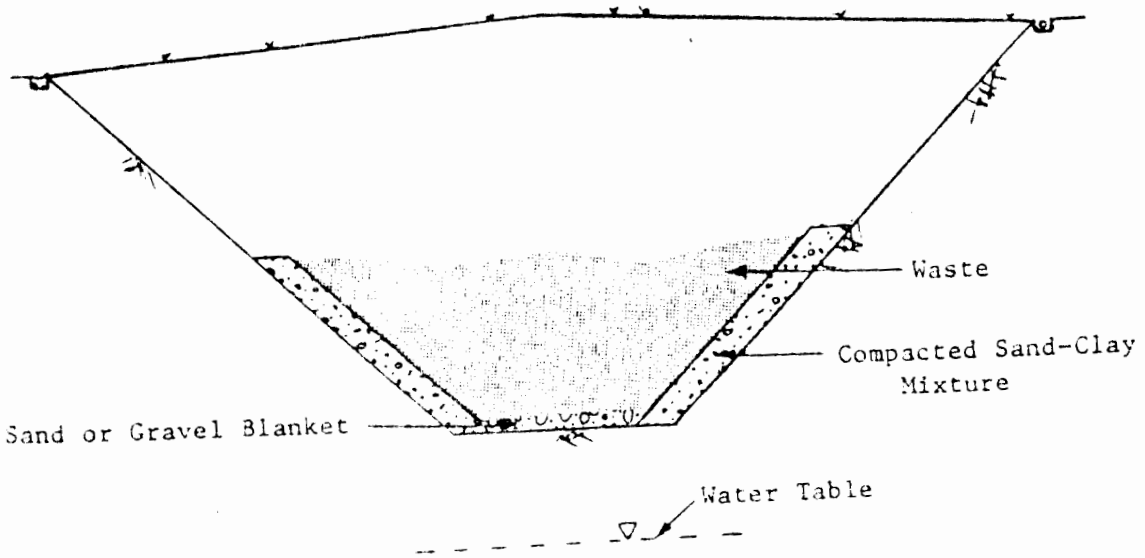
Liners could be used to retard the movement of radionuclides away from the disposal pit.

The liner must be nearly impervious and not susceptible to degradation by chemical reactions or radiation exposure. Compacted clay or sand-clay mixtures could be used because they are resistant to attack by many chemicals, and attenuate cationic radionuclides. Calcareous clays, however, should be avoided if the effluent is acidic.

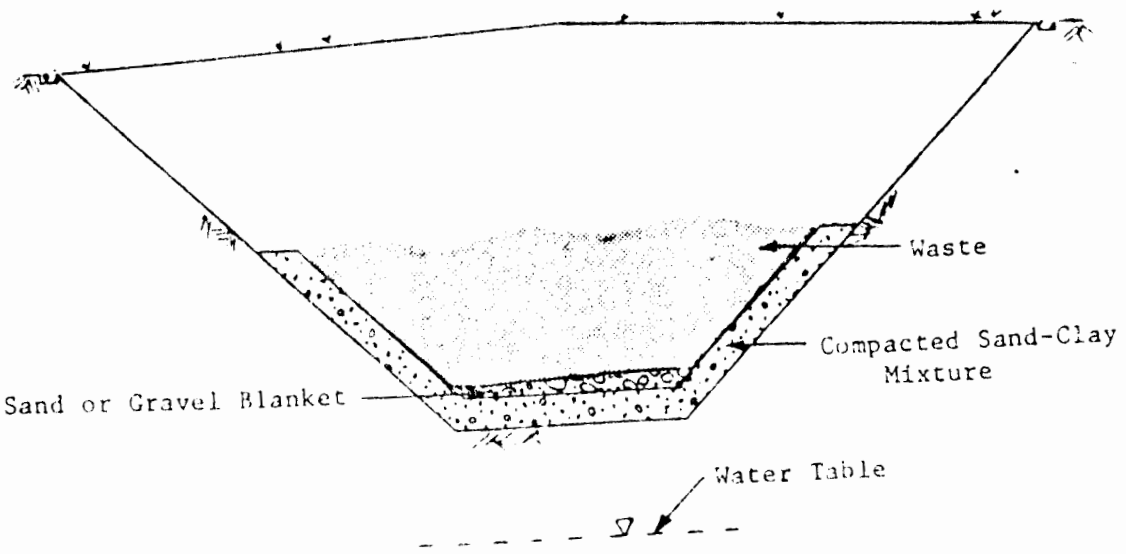
Lining the sides of the disposal pit (Fig. 2.4a) will minimize the possibility that radioactive leachate could migrate laterally. Whether to line the floor of the pit (Fig. 2.4b) may depend on several factors, such as the amount of precipitation and geology of the area. In essence, it would depend on whether a significant amount of contaminants would be released to the biosphere or whether dilution would mitigate future problems. If the bottom of the pit is lined, a collection system would probably be necessary. It might consist of a blanket of coarse-grained material on top of the floor of the pit, with a gentle slope to a sump area that is drained through one or more riser pipes.

Concluding Comments

Use of caps and liners is only one of various possible solutions for reducing some of the problems currently occurring at low-level radioactive waste disposal sites. For regions which do not have a high water table or a significant amount of precipitation, caps and liners should sufficiently retard the inflow of moisture and outflow of contaminants. However, for regions with a shallow water table where it may be necessary to bury the waste below the water table, additional barriers created by pretreatment of the waste (Sheppard, 1979) and/or use of containers specifically designed to retain the waste, may be necessary.



(a)



(b)

Fig. 2.4 Compacted sand-clay mixture used to line (a) only the sides of the pit, or (b) both the sides and bottom of the pit.

3.0

SHRINKAGE AND CRACKING OF COMPACTED CLAY LINERS

3.1 Introduction

Intact compacted clay liners would seem to offer great advantages for retarding flow into or out of disposal pits for wastes of many types and they are being used increasingly for this purpose. With thicknesses of a meter or more they are highly resistant to puncture. The constituent minerals are mainly silicate and thus tend to be resistant to attack by many chemicals. Local clays are used in most applications, thus minimizing costs. The major problem with compacted clay liners is the danger of cracking due to desiccation, or, less often, to wet/dry cycles or frost action. Cracking presumably occurs during a drying period when leakage of liquids would not be a problem but gases may be released during this time and there is a danger that the crack will not close completely during subsequent wet periods or will become filled with a more pervious material.

At the beginning of this study we made a strong effort to trace down references on shrinkage and cracking of compacted soils to determine the existing level of understanding. In spite of a wide recognition of the problem we found few useful references. It is possible that much of the published information consists of isolated sections of papers or reports on other topics and thus is difficult to retrieve. In this chapter we will review information found in the literature.

As a result of the lack of a well developed record of experience with shrinkage and cracking of compacted soils we began a series of preliminary

small scale laboratory tests to try to determine the main variables controlling shrinkage and cracking. The results of these tests are also included in this section.

3.2 Literature Review

It is well known that shrinkage cracks are absent in most compact clays of moderate to low plasticity, e.g., in glacial tills. The supposition is that some of these soils became dense because of desiccation but did not crack because the volumetric strain was too low. It may be noted that some glacial material became dense due to ice loading but dense clays in terminal, and recessional morains not later overriden by ice, must have been desiccated to achieve their high density. Desiccated deposits can be quite thick as evidenced by a 1 to 2 m thick crust in Chicago (Peck and Reed, 1954) up to a 7 to 10 meter thick crust in India (Kulkarni, et.al., 1975).

In considering the remaining natural clays that contain cracks and fissures, we are not concerned with fissures that have resulted from deposition and removal of overburden. These fissures appear to result from the fact that erosion of overburden tends to leave large lateral stresses locked into the soil. Continued erosion eventually reduces the vertical stress to the point that the ratio of the remaining lateral stress to the vertical stress is the critical value and failure occurs on sloping planes through the soil deposit (Brooker and Ireland, 1965).

Desiccation cracks tend to consist of nearly vertical tensile cracks. Ritchie and Adams (1974) observed the surface of Houston Black clay and reported shrinkage cracks to a depth of 0.90 m. Blight and Williams (1971) measured open shrinkage cracks to a depth of 0.65 m with closed cracks extending to 1.45m.

Sherard (1973) found shrinkage cracks up to 2 m deep in compacted clay in dams. We have heard rumors of significantly deeper shrinkage cracks but were unable to find actual data confirming deep cracking.

The mechanisms controlling cracking are not understood. It seems reasonably clear that cracking results from excessive volumetric strains during desiccation but we found no data on the magnitude of desiccation induced strain required to initiate cracking. A few direct measurements of tensile strengths and strains have been reported (Leonards and Narain, 1963; Krishnayya, Eisenstein, and Morgenstern, 1974; Ajaz and Parry, 1975). Tests were performed using direct tension, beam flexure, and indirect (Brazilian) tension. The data suggest that the tensile strain at failure increases as the water content increases, for any one soil, but otherwise few conclusions seem possible. Tensile strains at failure for the three studies ranged from 0.05% to 15% and appeared to vary with test type, loading rates, and undetermined soil characteristics.

The linear shrinkage in soil composed of parallel platey particles would be expected to be largest perpendicular to the flat surfaces of the particles (Lambe, 1960). If so, then in a soil with horizontally oriented platey particles, large amounts of vertical shrinkage might occur without formation of vertical cracks. Alternatively, soils with large tensile strengths might resist cracking by forcing strains in the vertical direction.

Corte and Higashi (1964) studied shrinkage cracking of small samples of clay prepared at water contents above the liquid limit and concluded that the water content at which cracking began was a function mainly of the rate of desiccation. The conclusion may not be applicable to compacted clays.

3.3 Laboratory Studies at the University of Texas

3.3.1 Working Hypotheses

The literature study provided little useful information on either the mechanisms controlling cracking or the extent of cracking in the field nor did it indicate useful methods of measurement in the laboratory. An ideal approach to the problem would have been to construct compacted fills in the field using a variety of clays, compacted at a range in water contents and densities, and observe cracking patterns and depths. As a result of the high costs and long duration of such tests it seemed desirable to perform small scale laboratory tests first.

An efficient testing program is organized by first developing a set of hypotheses or theories (the term depending on the existing knowledge) to explain the relevant phenomena, then developing specific experiments to prove or disprove the most critical parts of the hypotheses/theories. Successive refinement of measurements and theories should eventually lead to a useful solution. The dearth of available knowledge in this subject meant that we had to start with a set of hypotheses. These hypotheses were essentially as follows: Cracking results from shrinkage. Shrinkage is likely to be nearly equal in all directions in an unconfined sample. In a layer of soil, lateral tensile stresses will lead to vertical (normal to the flat surface) strains that constitute the entire volumetric strain until the lateral tensile stresses reach the tensile strength; then failure occurs and strains become more or less isotropic. During drying, the compression of the soil results from the increases in effective stress caused by negative pore water pressures. In the case of saturated soil, the effective stress ($\bar{\sigma}$) can be calculated from the total stress (σ) and the pore water pressure (u)

using Terzaghi's equation:

$$\bar{\sigma} = \sigma - u \tag{3.1}$$

The soil should shrink along a strain (ϵ) -effective stress curve ($\bar{\sigma}$) such as the one shown in Fig. 3.1 The soil shrinks to some limit, called the "shrinkage limit" that corresponds to effective stresses in clays of the order of 50,000 to 300,000 psf. ≈2.5 to 15 MPa For a given initial water content, the density (void ratio, porosity, etc.) at the shrinkage limit is a function of the soil structure, being higher when the platey clay particles are at large angles to each other and lower when they tend toward parallelism. For a given soil structure, with a unique shrinkage limit, the shrinkage strain should depend on the initial density - the higher the initial density the smaller the shrinkage strain and the less is the tendency to crack. For partially saturated soils, the effective stress cannot be defined uniquely. A widely used equation is (Bishop, et al., 1960).

$$\bar{\sigma} = \sigma - \chi u \tag{3.2}$$

where u is the pore water pressure (the pore air pressure is here taken as zero), and χ is a coefficient with values of zero for a completely dry soil and 1 for a saturated soil. If a soil is compacted at low water content, χ may be very low so that drying may induce little effective stress and thus little shrinkage. If the soil is first saturated, the increased value of χ will lead to greatly increased shrinkage on drying. Cyclic wetting and drying may produce a lower shrinkage limit, and thus more cracking, than simple drying because it would give soil particles more chance to slide into dense packing.

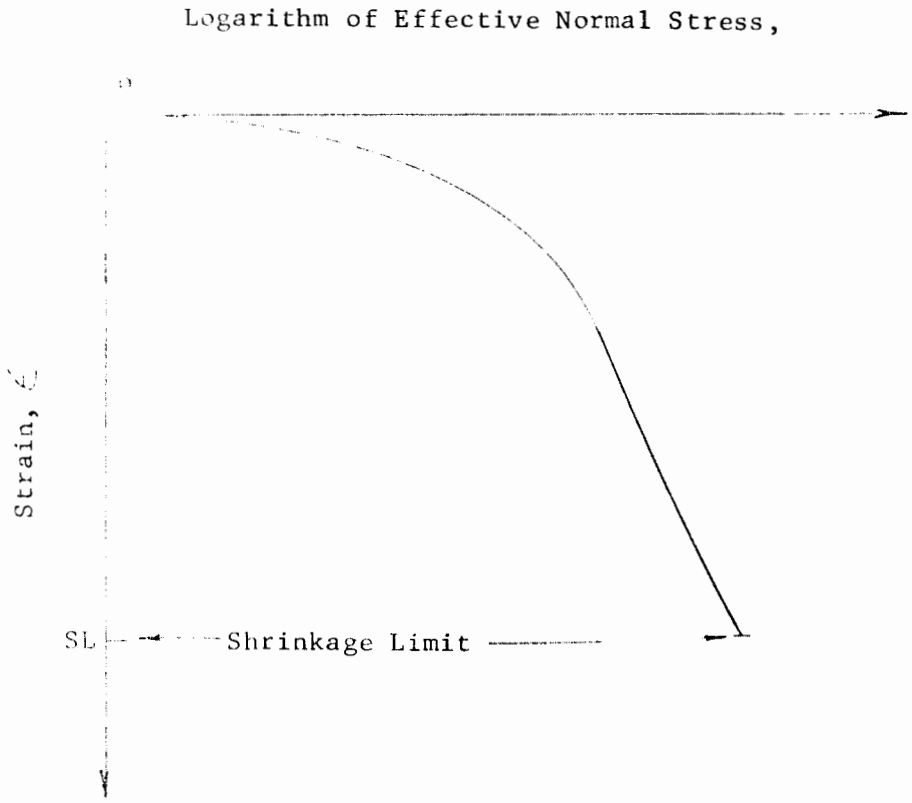


Figure 3.1 - Shape of the Strain-Effective Normal Stress Curve for Clays

A logical suposition would be that compaction to a high density would minimize volume change, and thus cracking. Compaction to the shrinkage limit should eliminate cracking. However, heavy compaction may tend to orient platey particles and thus lower the shrinkage limit. Also, cyclic wetting and drying may still lead to cracking because swelling is forced to be one dimensional whereas shrinkage tends to be three dimensional.

The problem is not just to minimize cracking, it is also to minimize permeability. Sands shrink very little upon drying, and do not crack but have a high permeability. Clays have reduced permeability but undergo increased shrinkage as plasticity increases, and often crack. A logical hypothesis is that a soil composed of a framework of sand particles with their interstices filled with clay, would have both low shrinkage and low permeability.

3.3.2 Types of Laboratory Tests

Two types of tests were used in this preliminary study. First, samples were compacted into metal molds, removed from the molds, and allowed to dry. Observations were made of sample weight, height, and diameter, as a function of time during air drying. After equilibrium was established the samples were oven dried and a final set of readings taken. Because of their small size and more-or-less equal dimensions, the samples did not crack. The main purpose of these tests was to determine the effects of soil type, and compaction water content and density on total shrinkage. A second set of experiments was performed on prismatic samples with the purpose of getting a qualitative idea of when shrinkage cracks would occur.

3.3.3 Tests with Cylindrical Samples

3.3.3.1 Sample Preparation

Tests were performed using two sizes of specimens. A number of tests were performed using samples compacted in a standard Proctor mold (4.0" I.D. x 4.58" high). This mold was chosen because of its wide use and because it can be used with natural soils containing particles up to say 1/4" in diameter. A larger number of tests were performed using a split mold, with 1.50" I.D. and 3" height, because less soil was needed, samples dried more rapidly, and samples were easier to handle. Samples prepared in Proctor molds were compacted in three layers, each struck 25 times using a 5.5-pound hammer (compaction energy of about 12,000 ft-lbs/ft^3). The smaller samples were compacted in ten layers using three to nine blows/layer of a 2.15-lb. hammer falling 12 inches (compaction energy ranged from 21,000 to 63,000 ft.-lb/ft^3). Two series of tests were performed initially, one using a local natural expansive clay, the so called Taylor clay (liquid limit=75%, plasticity index=49%, specific gravity of solids=2.73) and the other using a commercial fire clay, called the Elgin Fire clay (61, 41, 2.68).

3.3.3.2 Effect of Compaction Water Content on Shrinkage Strains

Measured values of dry density (at end of compaction), and measured strains in the radial (ε_{rad}) and axial (ε_{long}) directions, after oven drying, for samples of Taylor clay and Elgin clay, in the smaller mold, are shown in Figs. 3.2 and 3.3 respectively. In these plots, volumetric strains (ε_{vol}) were calculated from measured heights and diameters. The data show that the longitudinal and radial strains are nearly the same, indicating that the samples were nearly isotropic. The strains increased linearly with compaction water content. The data could be interpreted as indicating the desirability of compacting at very low water contents. However, if the

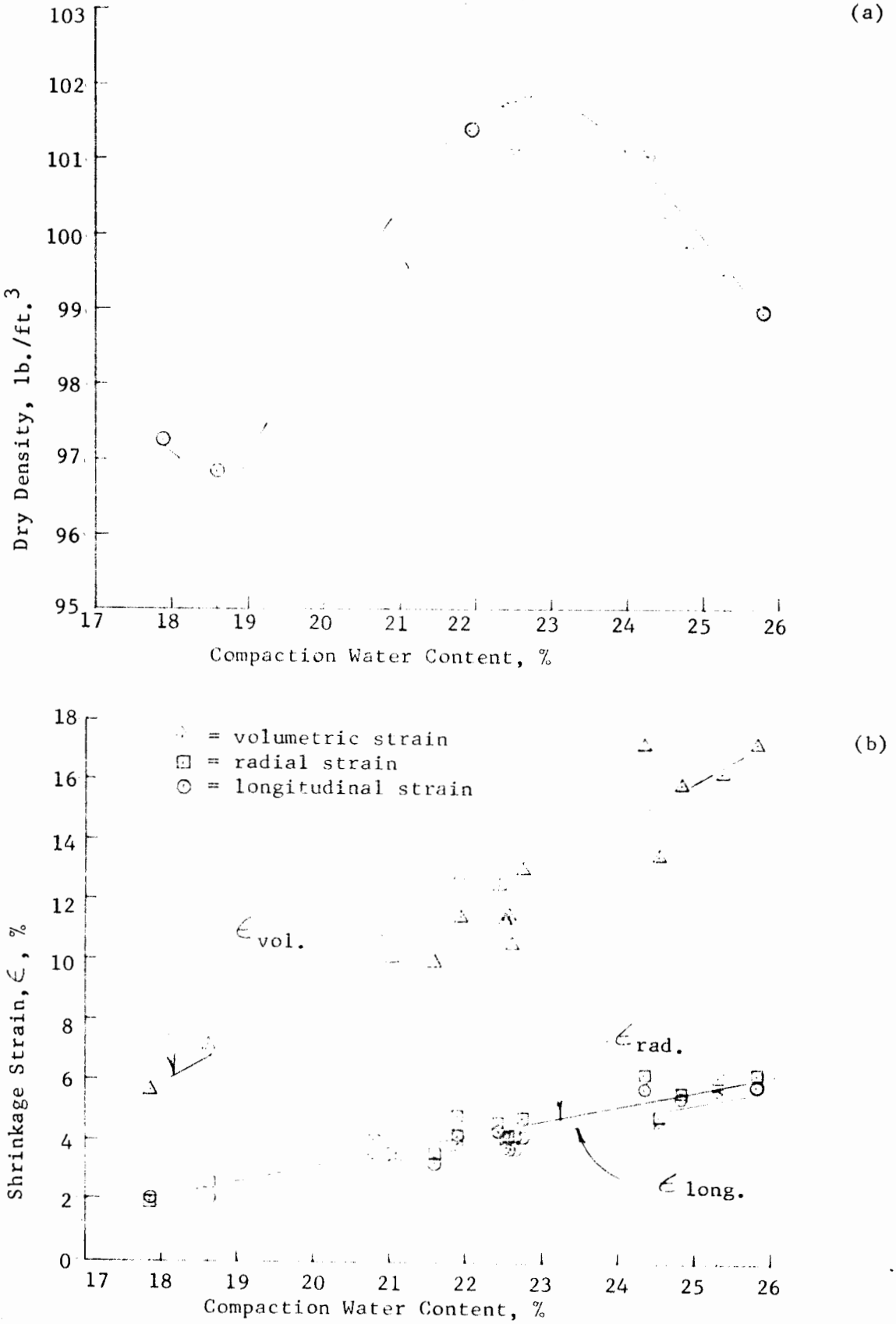


Figure 3.2 - Moisture-Density Relation and Shrinkage Strains for Taylor Marl -Small Cylinders -5 blows/layer.

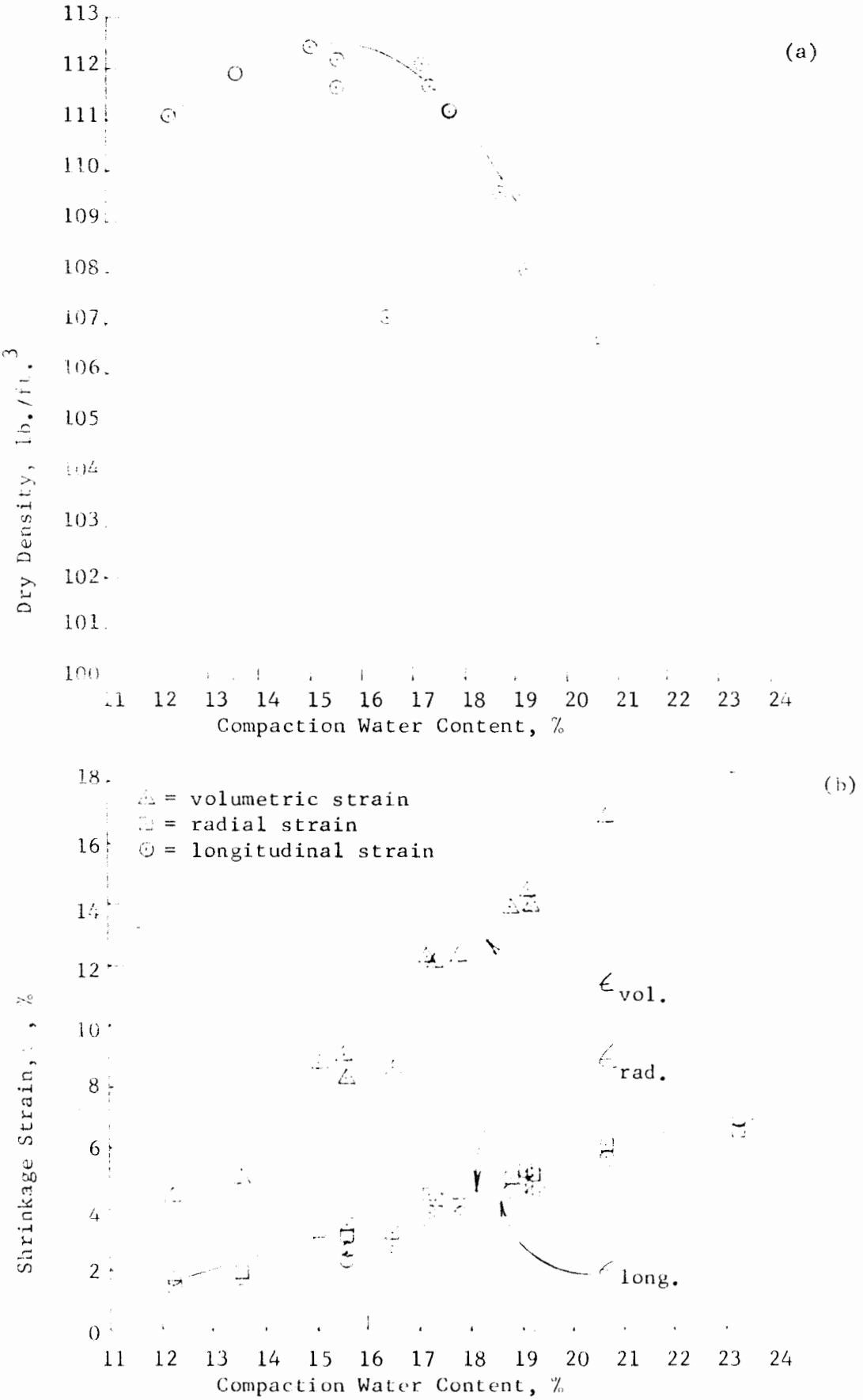


Figure 3.3 - Moisture-Density Relation and Shrinkage Strains for Elgin Fire Clay -Small Cylinders -5 blows/layer

samples were soaked first, as by rainfall in the field, we would expect the shrinkage at low compaction water contents to increase whereas samples compacted at high water contents should be unaffected. Tests on soaked samples will be performed next year.

3.3.3.3 Effects of Compacted Density on Shrinkage Strain

Surprisingly, the above tests did not yield reduced amounts of shrinkage for the densest samples. To study the effect of density further, samples were compacted in the small mold using 3, 6, and 9 blows/layer. Moisture density curves and shrinkage strains for Taylor clay and Elgin Fire clay are shown in Figs. 3.4 and 3.5, respectively. These data also confirm that for the range in compaction energies used here, dry density had a negligible effect on shrinkage strains, a fact in contradiction to our earlier hypothesis. A logical suposition is that, although heavy compaction reduces the as-compacted volume of voids it may also tend to disperse the soil structure, thus reducing the volume of voids at the shrinkage limit.

To save space, the individual plots for tests in standard Proctor molds will not be shown. The regression lines for tests using both mold sizes are shown in Figs. 3.5 and 3.7 for the two clays. Taken collectively the data suggest a slight tendency for shrinkage to increase slightly for increased compaction energy--a surprising result.

3.3.3.4 Effects of Coarse Particles on Shrinkage

To investigate the hypothesis that the presence of granular material will reduce shrinkage strains, shrinkage tests using the 1 1/2 x 3-inch samples were performed using mixtures of Los Alamos tuff, which is a silty medium to fine sand, and a commercial bentonite. Moisture-density curves for a range in bentonite fraction (in terms of air dry weights) from 12% to 100% are shown in Fig. 3.8. Shrinkage strains again varied more-or-less

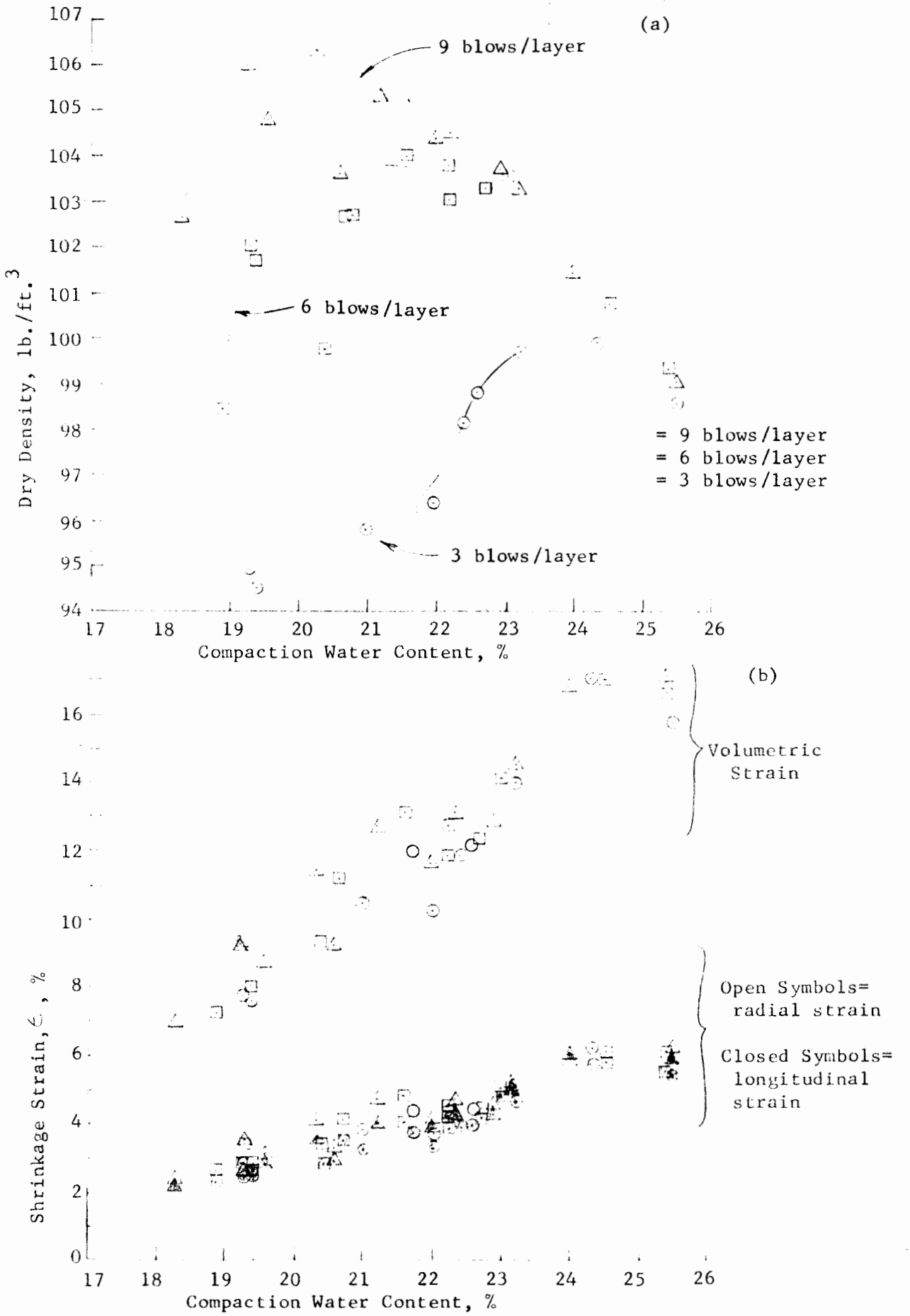


Figure 3.4 - Moisture-Density Relation and Shrinkage Strains for Taylor Marl -Small Cylinders -3, 6, and 9 blows/layer.

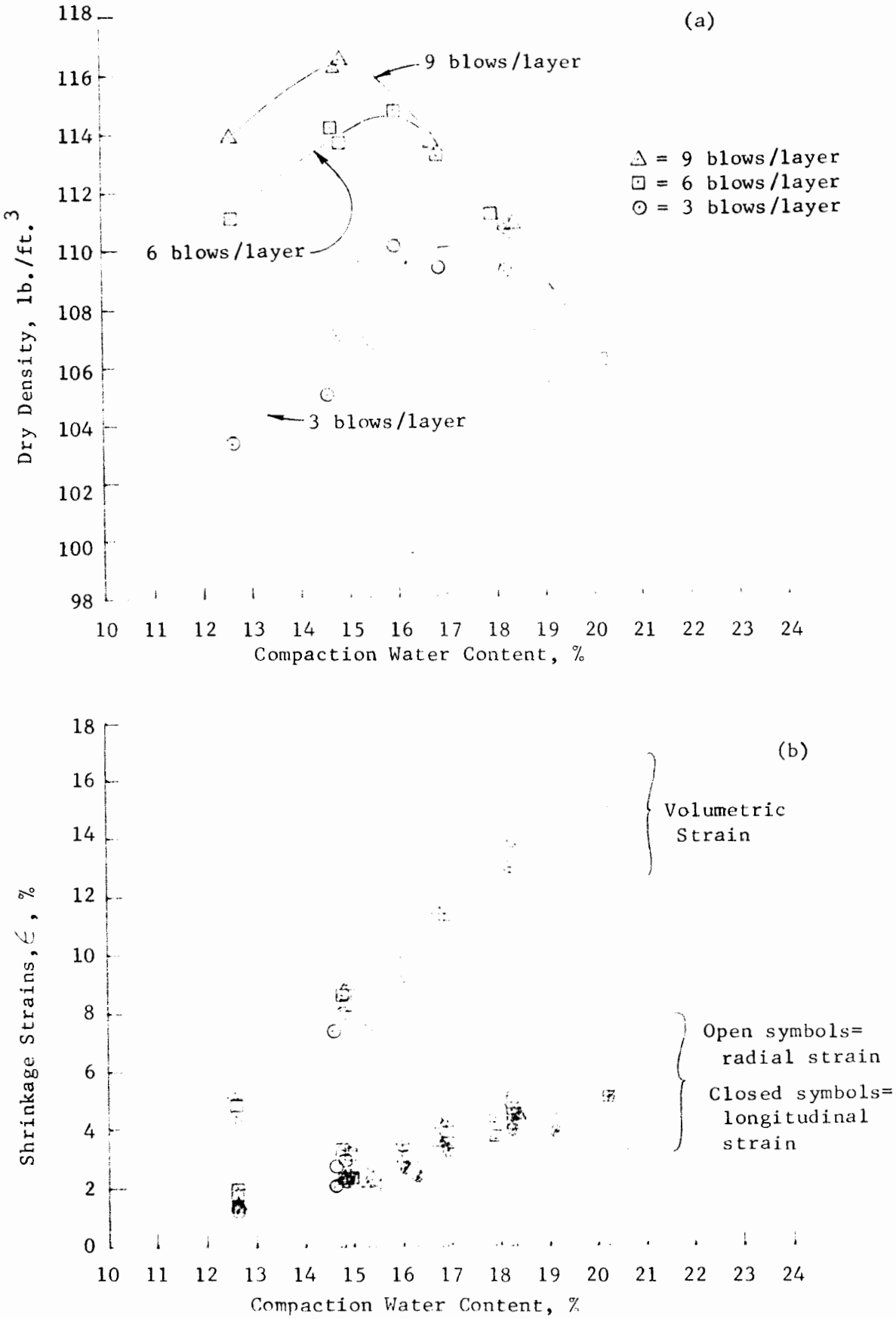


Figure 3.5 - Moisture-Density Relation and Shrinkage Strains for Elgin Fire Clay -Small Cylinders -3,6, and 9 blows/layer.

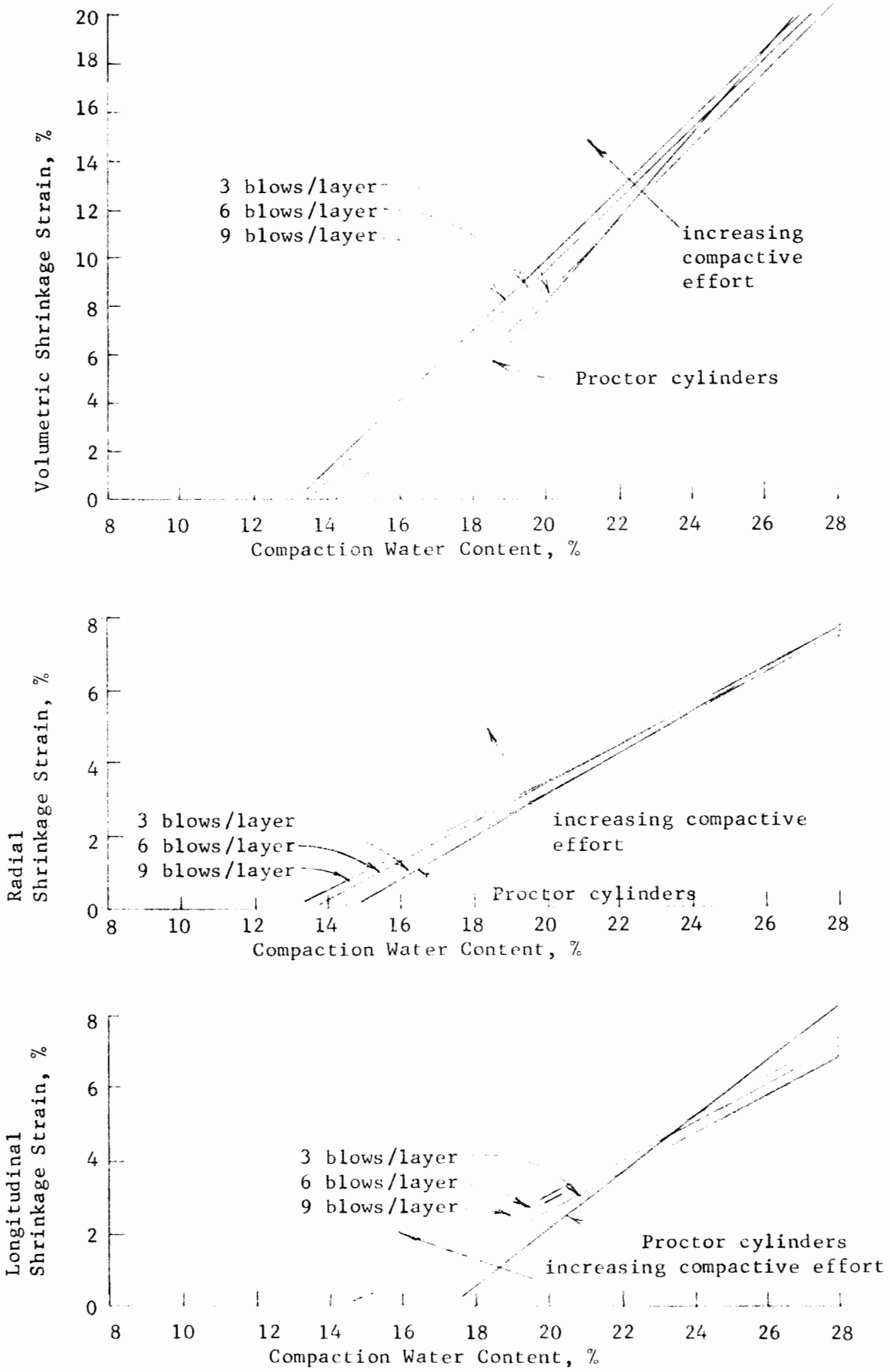


Figure 3.6 - Results of Linear Regression Analyses on Taylor Marl Shrinkage Tests

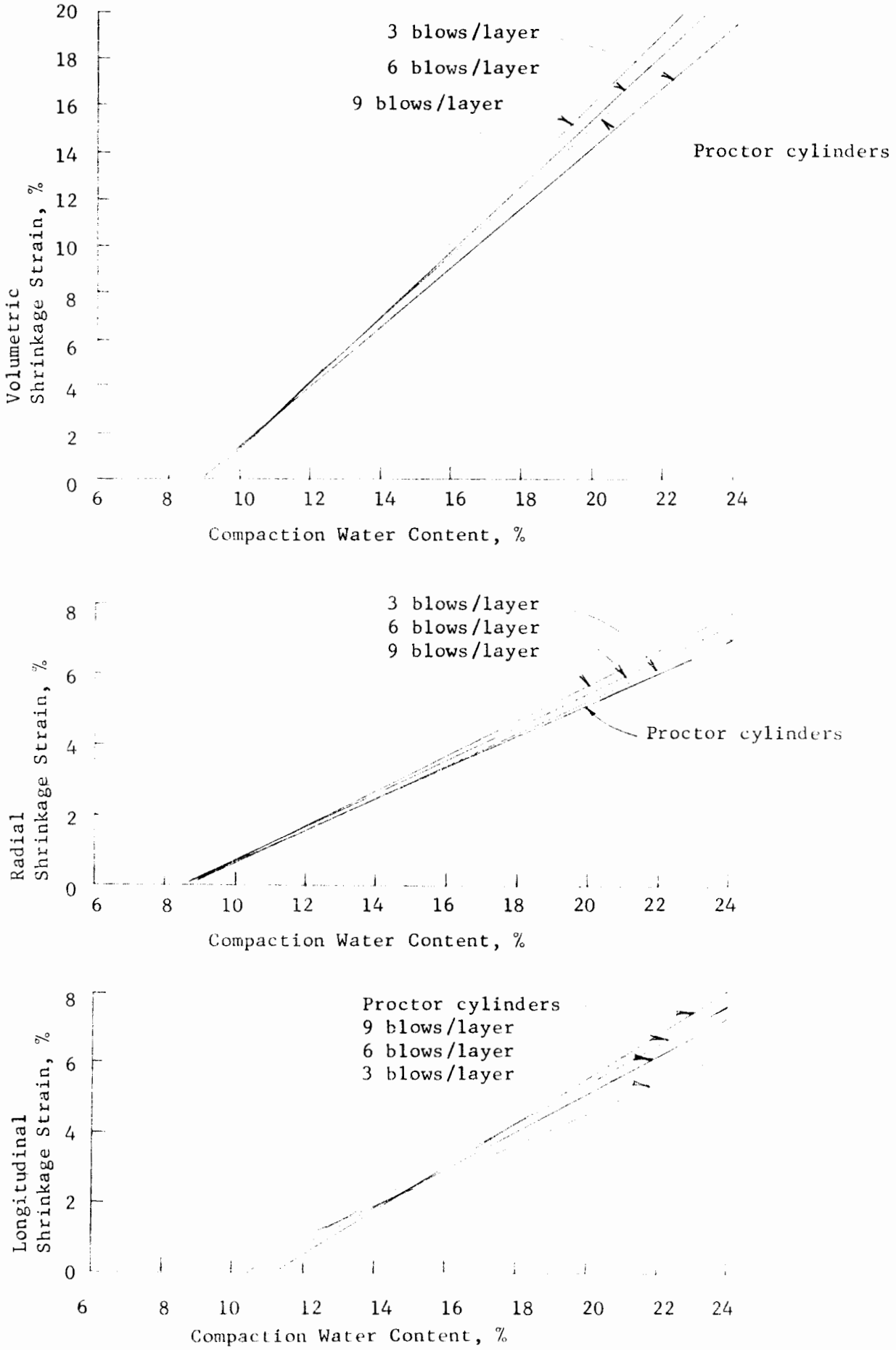


Figure 3.7 - Results of Linear Regression Analyses on Elgin Fire Clay Shrinkage Tests

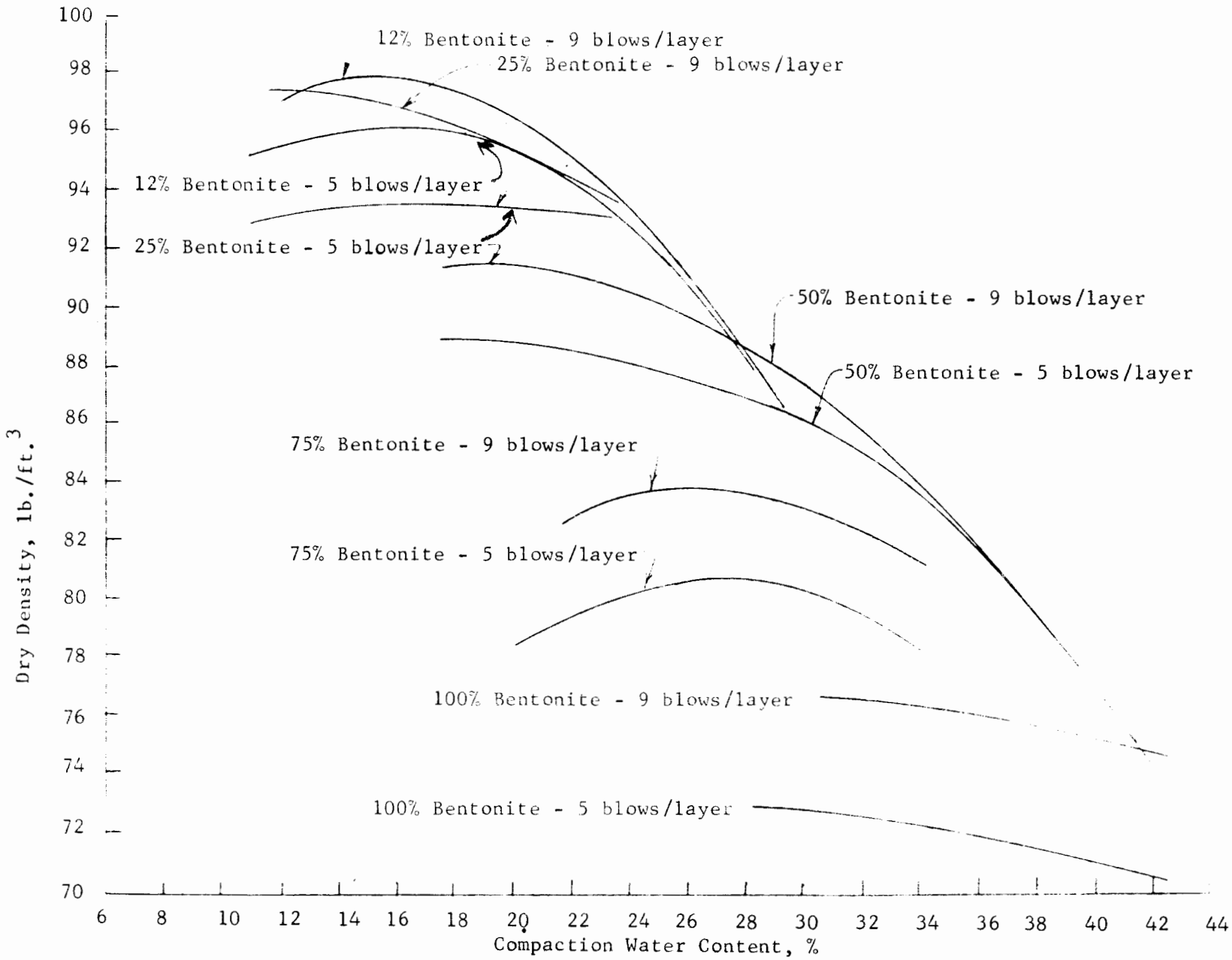


Figure 3.8 - Moisture-Density Relations for Los Alamos Tuff plus Bentonite Mixtures

linearly with compaction water content. The ranges in shrinkage and compaction water contents are summarized in Table 3.1. The ranges in water content are considered typical of what might be used in a field application. Negative strains indicate expansion. Permeability tests show that 12% bentonite renders the mixture relatively impervious (Daniel and Olson, 1980.) The tests appear to confirm the hypothesis that suitable soil mixtures can be made both crack resistant and relatively impervious.

3.3.4 Tests with Prismatic Samples

The data presented above provide indirect evidence of the probability of cracking. A more direct test was sought, preferably one that would simulate field conditions. The critical field condition would consist of a wide relatively thin layer of soil drying from the surface down. The base of the layer would not deform because it would be in intimate contact with unshrunk soil. As a practical matter, wide thin samples cannot be used for general testing. We chose to use brick shaped samples. To restrain the base, compacted samples were placed on a plastic plate with holes in it. Thin steel nails were pressed up through the holes about 1/2 inch into the soil to prevent shrinkage strains parallel to the plate. Initially, a series of samples of Taylor clay and Elgin Fire clay were compacted using standard Proctor energy (12,375 t-lbs/ft^3) with a thickness of 3 inches and horizontal dimensions of 6" x 12". All samples compacted dry of optimum cracked upon drying. All samples compacted wet of optimum curled up by lifting the ends off the nails. Another set of samples was prepared with a thickness of 2 inches but they behaved similarly. The apparatus was redesigned to provide hold-down clamps at the two ends. All samples provided with hold-down clamps cracked. It should be noted that all of these tests utilized either

Table 3.1 -- Shrinkage Strains for Los Alamos Tuff and Bentonite Mixtures

Percent Bentonite	No. of Blows per Layer	Range of Water Content (%)	Range of Shrinkage Strains (%)		
			Volumetric	Radial	Longitudional
12	5	11 - 28	-.1-3.0	0-0.2	-0.2-1.2
12	9	12 - 29	0.1-4.0	0.1-0.3	-0.3-1.6
25	5	11 - 23	1.2-5.4	0.6-1.2	0-3.3
25	9	12 - 23	1.2-3.4	0.6-1.5	0-0.5
50	5	17 - 42	3.7-15.2	1.3-5.6	1.1-4.9
50	9	18 - 41	3.6-15.6	1.6-6.2	1.1-4.0
75	5	21 - 34	8.1-18.8	2.7-7.1	3.0-5.9
75	9	22 - 34	8.6-18.0	2.9-6.3	3.1-6.5
100	5	28 - 42	18.0-30.3	6.5-11.2	6.2-11.7
100	9	31 - 42	20.5-32.2	7.4-12.1	7.3-12.3

the Taylor clay or the Elgin Fire clay, both of which are highly susceptible to cracking.

Observations were made of sample weight vs time and of the time of appearance of cracks. First cracks appeared within 24 hours. The average water content at the time of cracking (24-hour reading) is plotted versus compaction water content in Fig. 3.9. Apparently, a loss in average water content of about 2% led to the onset of cracking. The diagram does not indicate the severity of the cracking, which actually varied from minor surface cracking for the samples at lowest water content to deep cracks for the samples at the highest water content.

3.4 Summary and Conclusion

Although potential problems with cracking of compacted clays are widely known and discussed, little research appears to have been performed to determine the mechanisms that cause cracking and no methods exist either for predicting crack depths and spacing nor for performing diagnostic tests for cracking potential.

Preliminary tests reported here indicate that shrinkage caused by monotonic (as opposed to cyclic) drying increases linearly with compaction water content and is little affected by compaction density. No tests were performed using presoaked samples but it is believed that presoaked samples compacted at low water content and then dried will undergo more shrinkage than samples dried without soaking.

Based on one set of tests it appears that soils can be prepared (if not found naturally) that contain sufficient coarse material to prevent shrinkage and cracking but sufficient fine material to reduce the permeability to an acceptably low-level.

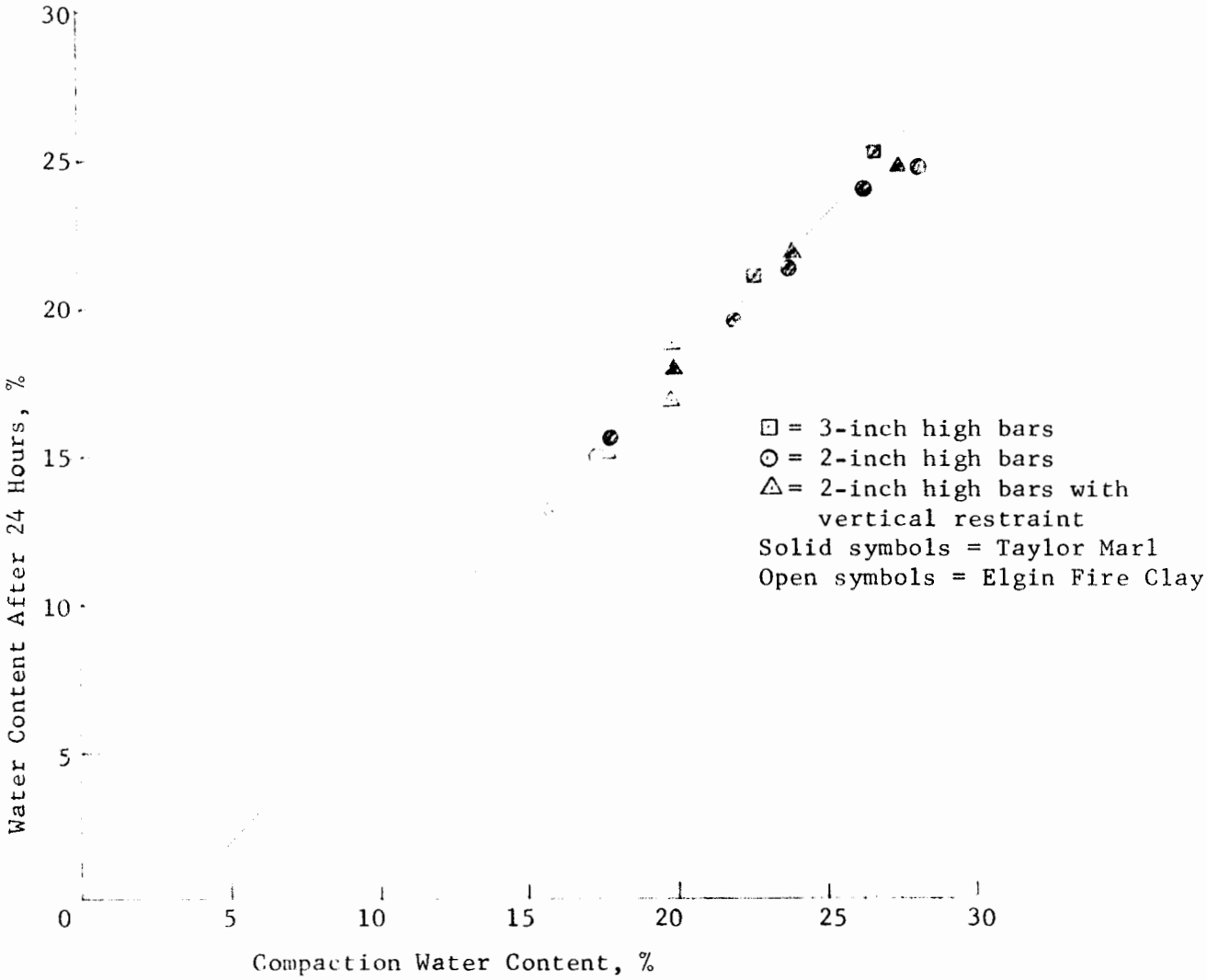


Figure 3.9 - Water content after Twenty-four Hours Versus Compaction Water Content for Compacted Soil Bars

Future tests include cracking studies on clay-sand and tuff-bentonite mixtures. Cylinder shrinkage tests on presoaked samples are also planned.

4.1 Introduction

The engineer or geologist who analyzes and designs a low-level waste disposal facility will be expected to provide some indication of how long it will be before pollutants begin escaping from the disposal pits, what the rates of escape will be, and what the concentration of pollutants will be in the escaping fluid.

The problem of making such predictions practically defies analysis because of the complexity of the problem. Factors to be considered in analysis include meteorology, vegetation, properties of the cap, concentration of radioactivity in the waste, chemistry of the waste, solubility of radionuclides in water, properties of the liner, and properties of the subsurface soils or rocks. In addition, a correct analysis would have to include effects of flow due to temperature gradients, two-phase flow in unsaturated porous media, chemical interactions between dissolved pollutants and the soil, and volume change as the soil is wetted or dried.

The basis for any prediction of field performance must be a model for water flow in unsaturated soil, with as many of the effects noted above included within the capabilities of the model as is practical and desirable. Work in previous phases of this project led to development of a computer program called FLOW, and later FLOW4, for analysis of moisture movement in saturated or unsaturated soil. Work during this phase included expanding the program to analyze flow in two dimensions and to include some improved features.

4.2 Theory

For incompressible soil and pore water, Darcy's law may be combined with principles of conservation of mass to derive the following differential equation for isothermal flow of fluids in unsaturated porous media:

$$c \frac{\partial h}{\partial t} = -\nabla \cdot (k \cdot \nabla h) \tag{4.1}$$

where c, h, and t represent the slope of the moisture characteristic curve ($c = d\theta/dhp$, where θ is volumetric water content and hp is pressure head), total head, and time, respectively, and k is a matrix of hydraulic conductivities that may be written as follows for two-dimensional problems in cartesian coordinates:

$$k = \begin{bmatrix} k_{xx} & k_{xy} \\ k_{xy} & k_{yy} \end{bmatrix}$$

and ∇ is the operator $\partial/\partial x + \partial/\partial y + \partial/\partial z$.

4.3 Computer Programs

Equation 4.1 is highly non-linear because the slope of the moisture characteristic curve and the hydraulic conductivity may change by several orders of magnitude over the range in saturation of interest in practical problems. The non-linearity, coupled with complex geometry and boundary conditions, makes closed-form analysis impossible. A number of finite element and finite difference computer programs have been written to analyze the problem but all suffer from deficiencies. The most important deficiency is that a majority of existing programs were written by agronomists whose interest was in infiltration of water into the ground; such programs typically

consider only one-dimensional flow of water. The two-dimensional finite element codes are the most suitable ones, but only a few programs have received significant national attention. The program by Reeves and Duguid (1975), called SOGMOD, was among the first programs to be written that was capable of analyzing two-dimensional problems with realistic boundary conditions. Unfortunately, the code was not well suited to deal with the highly nonlinear nature of the governing differential equation and was not recommended for use in unsaturated soils. A code described by Zyvoloski et al. (1975) is similar to Reeves and Duguid's program in many respects, but seems to contain more restricted options for modelling boundary conditions and soil properties. Frind et al. (1979) have developed a two-dimensional finite element code, but it offers few refinements and was apparently developed mainly for use in analyzing results from laboratory tests. The program originally developed by Baca et al. (1978), called SEMTRA, is still under development and probably represents the most sophisticated program available. However, the program is so complex that we are told running times for a complete analysis of a problem os significant size are typically 1 to 2 days on a Univac model 400 computer. This program is probably more sophisticated than can be justified considering the quality of data that would be fed into it.

We have been developing a two-dimensional finite element code for analysis of water flow in saturated or unsaturated soils. In the program, emphasis is placed on convenience of use, accuracy and flexibility of calculations, and generation of meaningful output. The program is called DUNSAT1 and is described briefly below.

4.4 Program DUNSAT1

Program DUNSAT1 analyzes isothermal movement of water in saturated or unsaturated soils. The methods of solution is a Galerkin approximation (Zienkiewicz, 1977) of the space derivatives and either a backward difference or trapezoidal rule for time integration (Zlateve and Thomsen, 1979). The program can be used to analyze one-dimensional problems, two-dimensional problems in cartesian coordinates, or two-dimensional problems in cylindrical coordinates. Other features of the program include the following:

- Nodal point coordinates and the finite element grid are generated internally.
- Four types of elements are possible: triangles or quadrilaterals with either linear or quadratic shape functions.
- Dynamic dimensioning permits use of minimum computer storage and allows analysis of virtually any size of problem without having to adjust size of dimensioned variables.
- Relevant data are input directly rather than being modelled with simplified and analytic functions. Most other programs have analytic functions built into them to model soil properties, but these functions sometimes do not work well, particularly for relative relatively dry soils. Program DUNSAT1 interpolates between input data points either on a natural scale or on a log-log scale.
- Several types of boundary conditions are allowed: precipitation with provisions for runoff; prescribed flux; prescribed total head, pressure head, or suction; freely draining boundary; and impervious boundary.
- Time step is controlled internally.
- Analysis of imaginary tracers is possible. An imaginary tracer is analagous to a drop of dye placed in the pore water at a particular point; the program will track the movement of the dye with time and print the coordinates of the tracer at all output times. This permits the user to track precisely the theoretical movement of water through the medium of interest.
- Both saturated and unsaturated flow can be analyzed. A majority of programs are formulated in such a way that saturated flow is not permitted.

The major limitations of the program are that isothermal conditions are assumed, hysteresis in soil behavior is not included, and movement of moisture in the vapor phase is not considered. The program also does not account for chemical interactions, e.g., ion exchange, because such interactions tend to be complex and to account for them would increase computer costs significantly. A conservative approach would be to design the disposal installation on the basis of predicted rates of water movement and to recognize that many dissolved pollutants in the water will move slower than the water.

4.5 Analysis of an Example Waste Disposal Installation

In this section, results of an analysis of a hypothetical waste disposal installation are presented to provide some indication of how the computer program might be used for a practical problem.

4.5.1 Site Conditions

The site under consideration is assumed to be located in a semi-arid region where, over the long term, more moisture is removed from the ground by evaporation than is added by precipitation.

The subsoil consists of a deep deposit of silty sand whose properties are assumed to be similar to Los Alamos tuff. The groundwater table is located 110 ft below the surface and is believed to have remained at that elevation for at least the past several hundred years. The groundwater below a depth of 110 ft is a valuable resource in the area. One design criterion is that no pollutants can reach the groundwater table in the first 100 years of operation of the disposal facility.

It is assumed from the outset that the pit will be lined. Clay is available locally, and a 5-ft-thick liner is tentatively planned. The

liner will be compacted dry of optimum to minimize formation of desiccation cracks during construction. The cap will be constructed of sandy clay that will also be compacted dry of optimum.

The geometry of the pit, liner, and cap is illustrated in Fig. 4.1. Soil properties used in the calculations are indicated in Figs. 4.2 and 4.3, and in Table 4.1. To simplify interpretation and presentation of results, all materials were assumed to be incompressible. The finite element grid used to analyze the problem is shown in Fig. 4.4 and consists of 273 linear elements with 301 nodal points.

Initial suctions in the silty sand were calculated by analyzing a one-dimensional column of soil in which the water table was fixed at a depth of 110 ft and a flux of -0.2 cm/yr (a negative flux indicates evaporation) was imposed at the ground surface. An initial distribution of suction was assumed, and the run was continued until steady-conditions developed. The computed initial profile of suction is shown in Fig. 4.5. The computer run to determine these initial conditions required 18 sec of TM time on a Cyber 150/750 computer.

Initial suction in the cap and liner was assumed to be 10 atm. The waste was assumed to be saturated initially, with a suction of zero.

4.5.2 Boundary Conditions

Boundary conditions for analysis of the disposal pit are shown in Fig. 4.6. No net evaporation was assumed to take place through the cap, on the assumption that the cap would be irrigated with small to moderate amounts of water in order to maintain a vegetative cover for erosion protection.

4.5.3 Results

The run was carried out to a calculation time of 100 yrs, with data

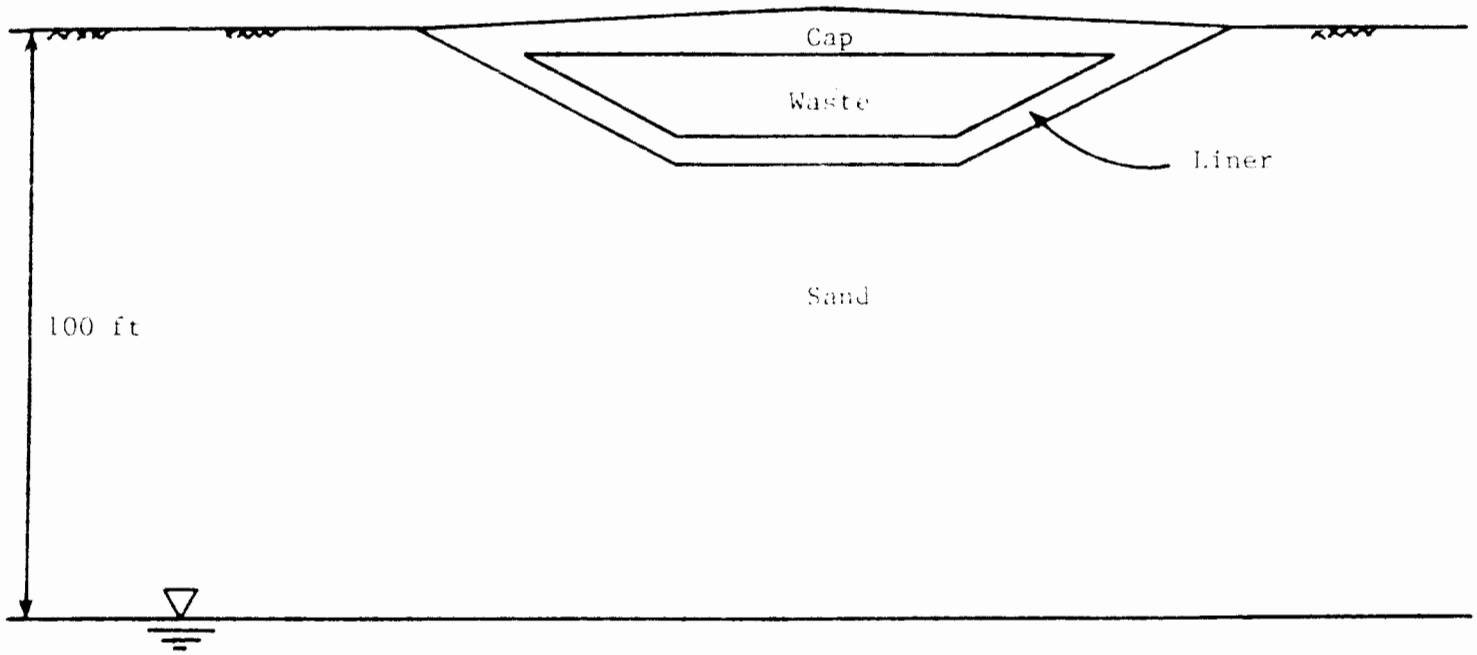


Fig. 4.1 -- Geometry of the Disposal Pit.

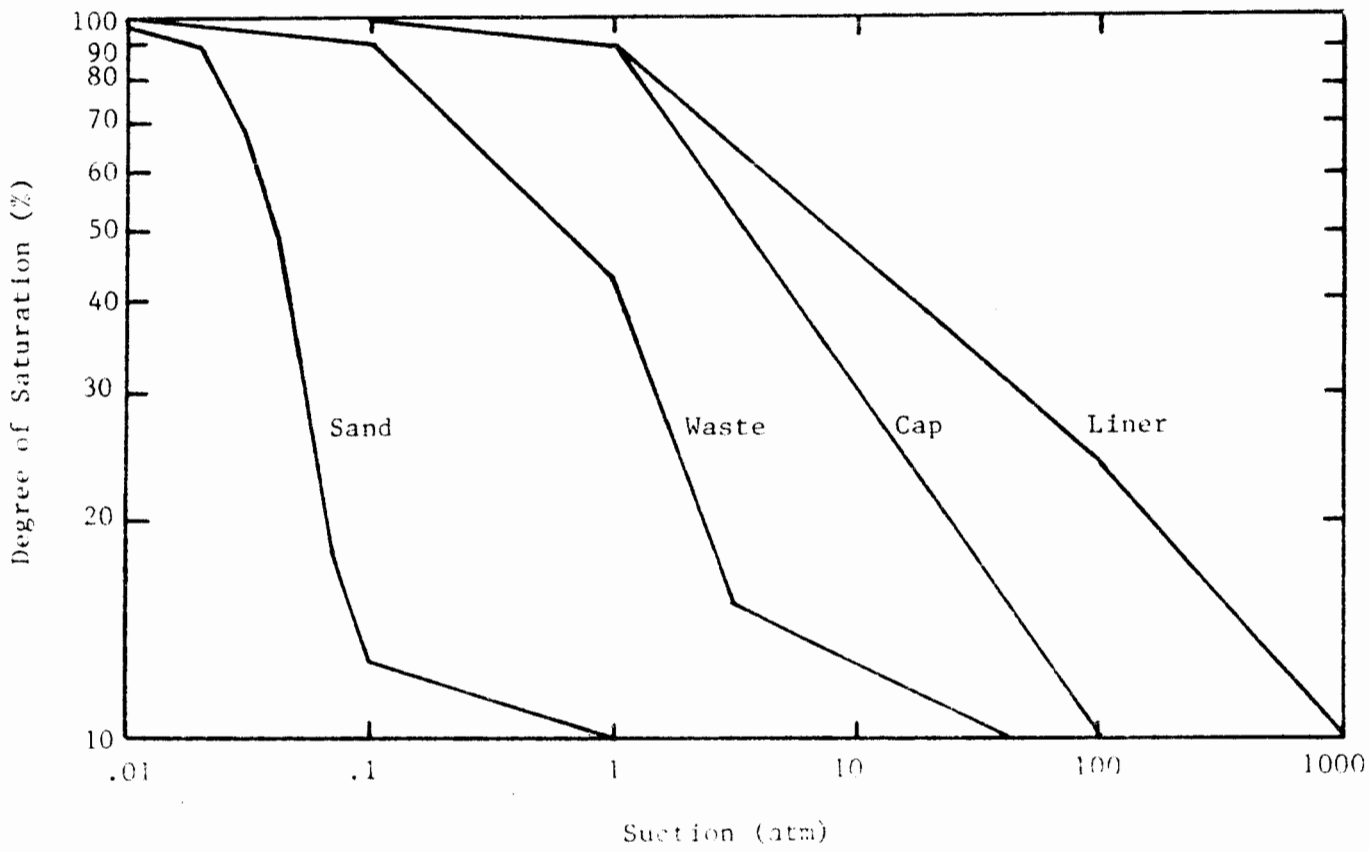


Fig. 4.2 -- Soil Moisture Characteristic Curves.

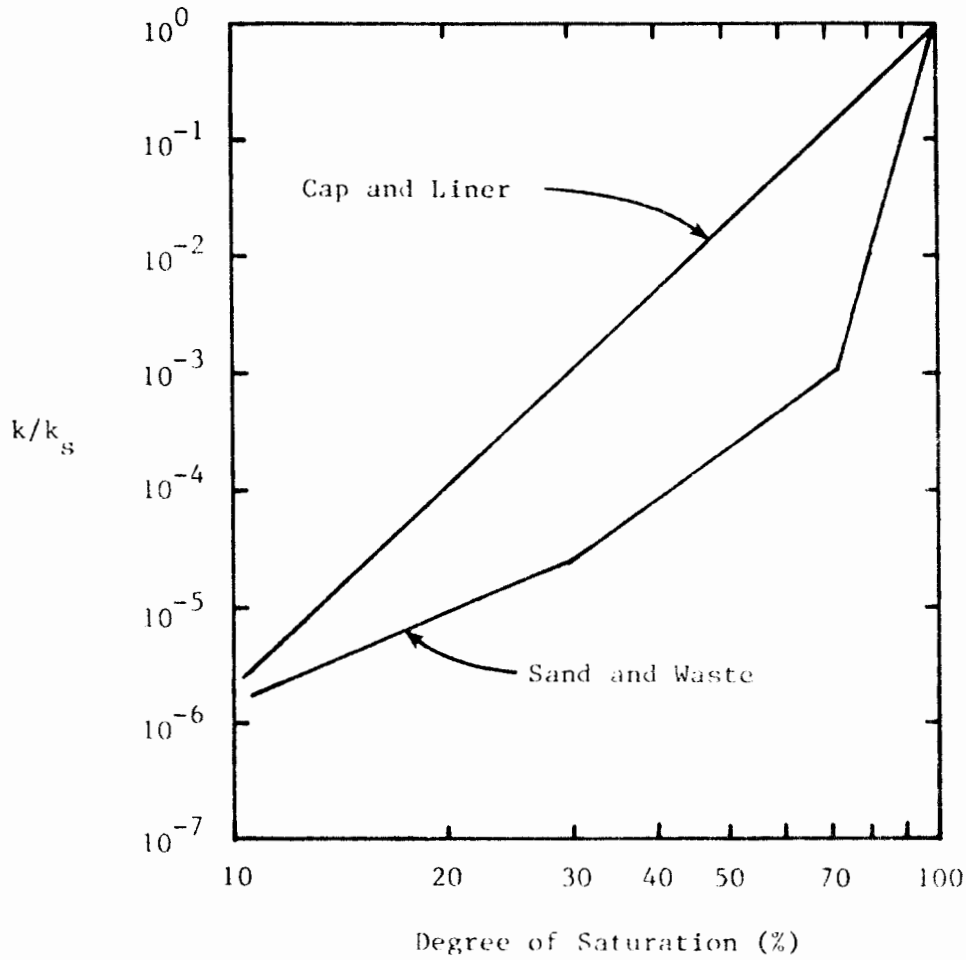


Fig. 4.3-- Hydraulic Conductivity Functions Used for Analysis.

Table 4.1

Soil Properties

Material	G_s	γ_d (g/cc)	n	n_e	Saturated Conductivities (cm/sec) k_{xx}	k_{yy}	k_{xy}
Cap	2.54	1.51	.41	.41	1×10^{-7}	1×10^{-8}	0
Waste	2.00	.40	.80	.80	1×10^{-2}	1×10^{-2}	0
Liner	2.70	1.70	.37	.37	1×10^{-7}	1×10^{-8}	0
Sand	2.50	1.51	.40	.40	1×10^{-4}	1×10^{-4}	0

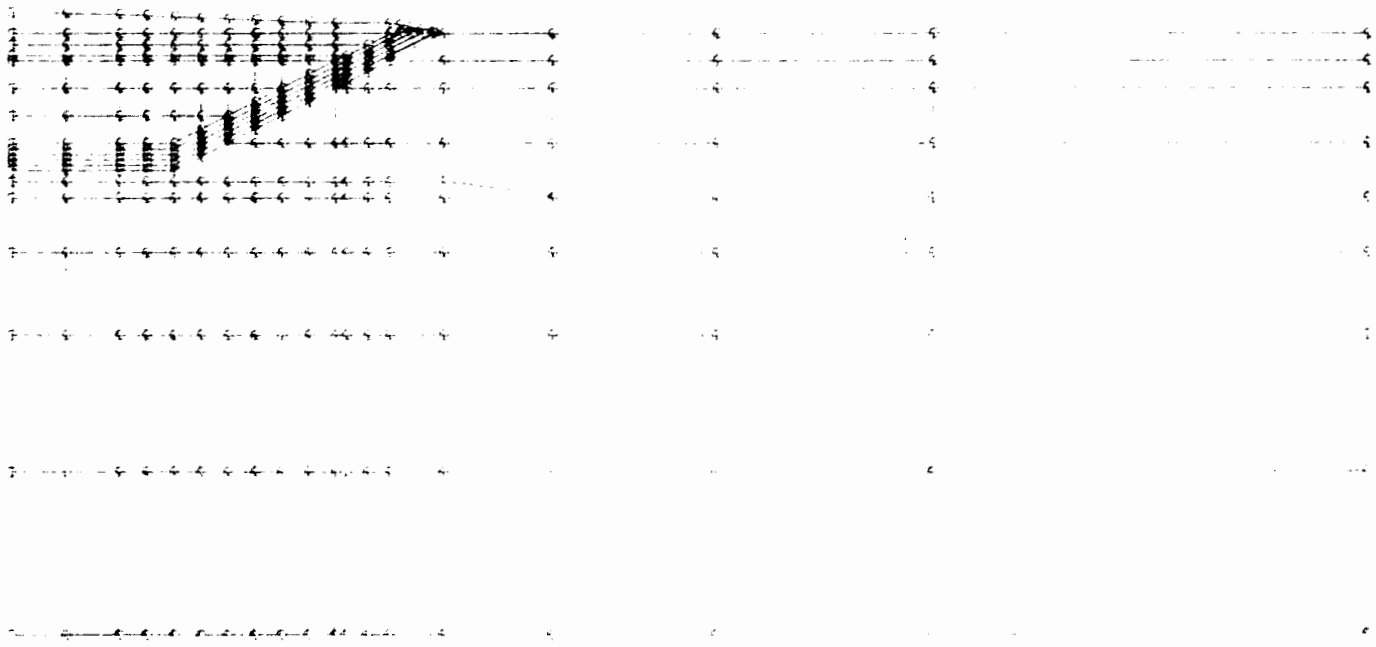


Fig. 4.4-- Finite Element Grid.

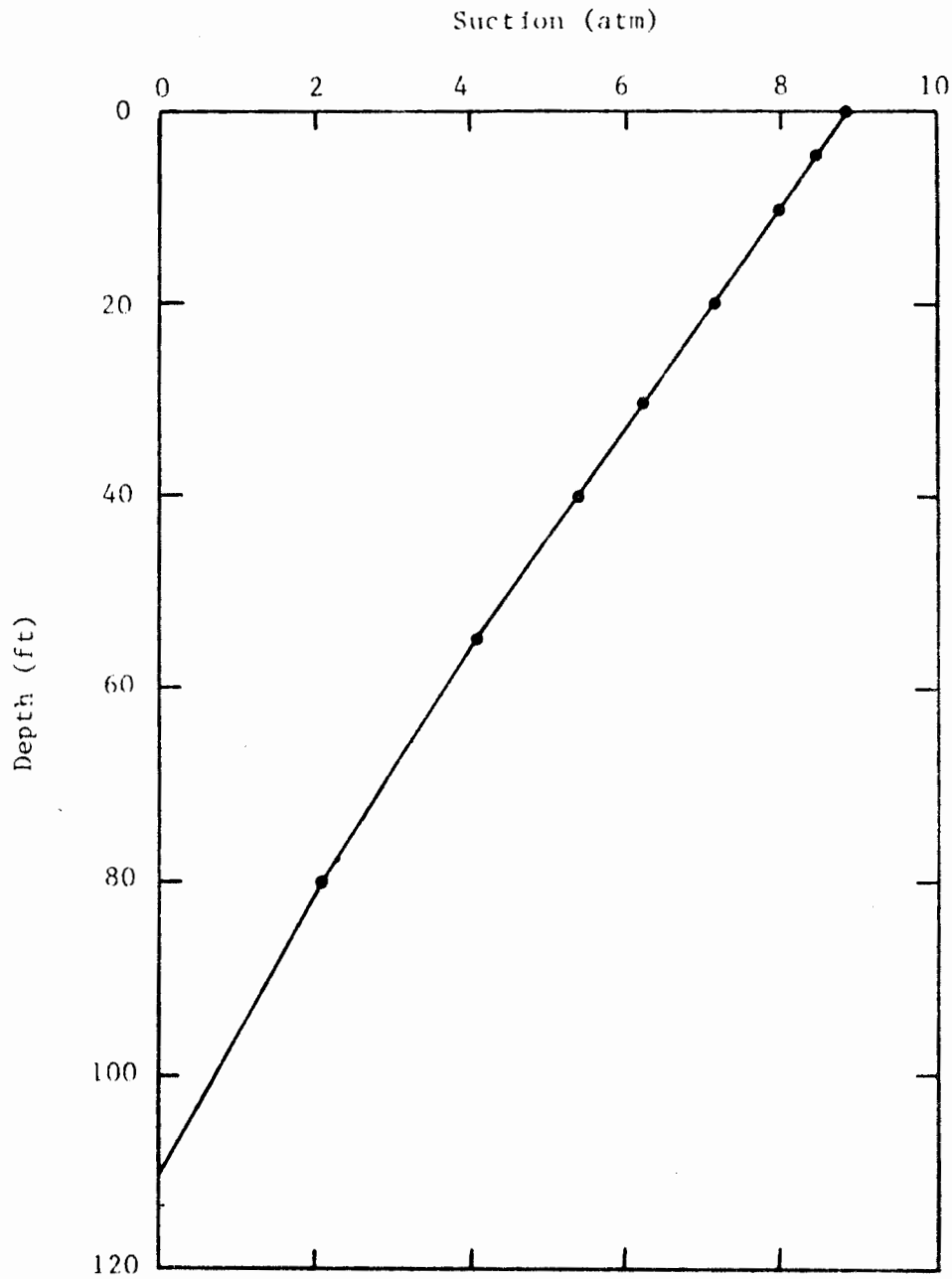


Fig. 4.5 -- Initial Suction Profile.

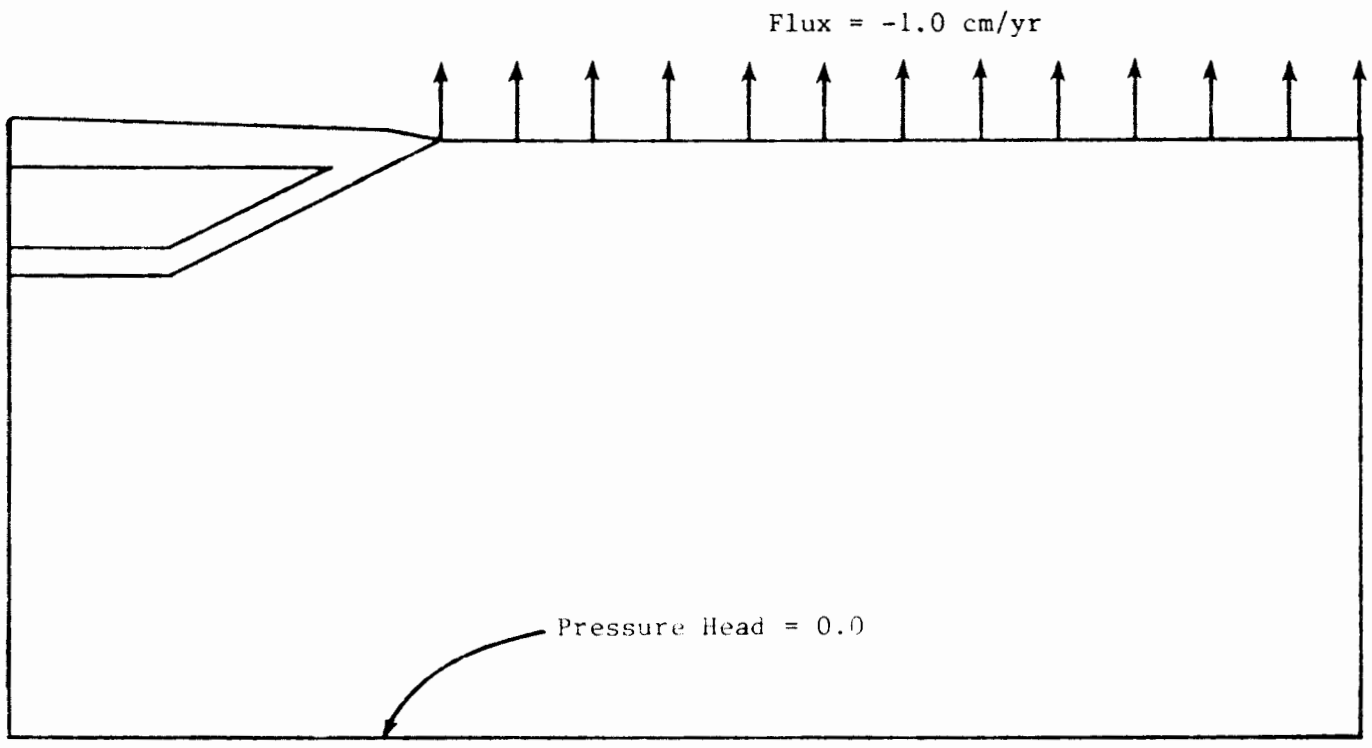


Fig. 4.6-- Boundary Conditions.

being output at calculation times of 0.1, 1, 5, 10, 20, 50, 75, and 100 yrs. The analysis required 32,000 words of storage, 89 time steps, 184 total iterations, 635 sec of central processor time, and 1031 sec of TM time on a Cyber 170/750 computer. Total cost of the run was $66, not counting plotting costs.

It is convenient to begin with interpretation of the results by comparing the change in suction with respect to time at several points (Fig. 4.7). Just beneath the liner, at point A, the suction initially increases slightly, because the suction is higher in the liner than in the underlying silty sand and therefore water flows from the sand into the liner. After the initial rise in suction, the suction drops steadly for about 25 years and then remains fairly constant. Examination of the fluxes at this point indicates that water is flowing upward, which means that the drop in suction is caused by water flowing upward from beneath rather than downward from the waste. The upward flow of water in the area below the pit is caused by cutting off evaporation from the area of the pit (Fig. 4.6). This idea is illustrated more clearly by coutours of constant suction shown in (Fig. 4.8).

At point B (Fig. 4.7), which is in the waste, the suction gradually rises with time in response to outflow of water. At point C, in the middle of the liner, suction drops rapidly at first, as the liner is moistened from water flowing out of the waste, but later follows essentially the same trend set by the point in the waste. Point D (Fig. 4.7) is relatively unaffected.

These same ideas can be illustrated by examining the change in degree of saturation at various points (Fig. 4.9). Degree of saturation drops

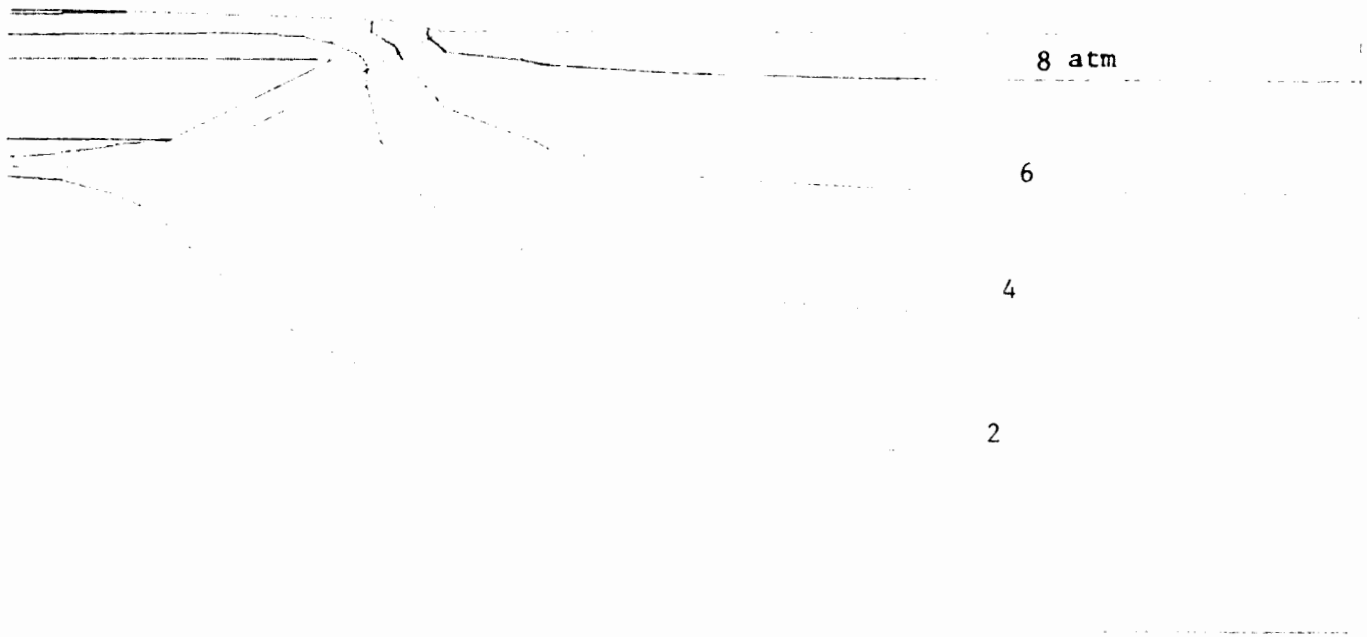


Fig. 4.7 -- Contours of Constant Suction at

A Calculation Time of 100 yrs.

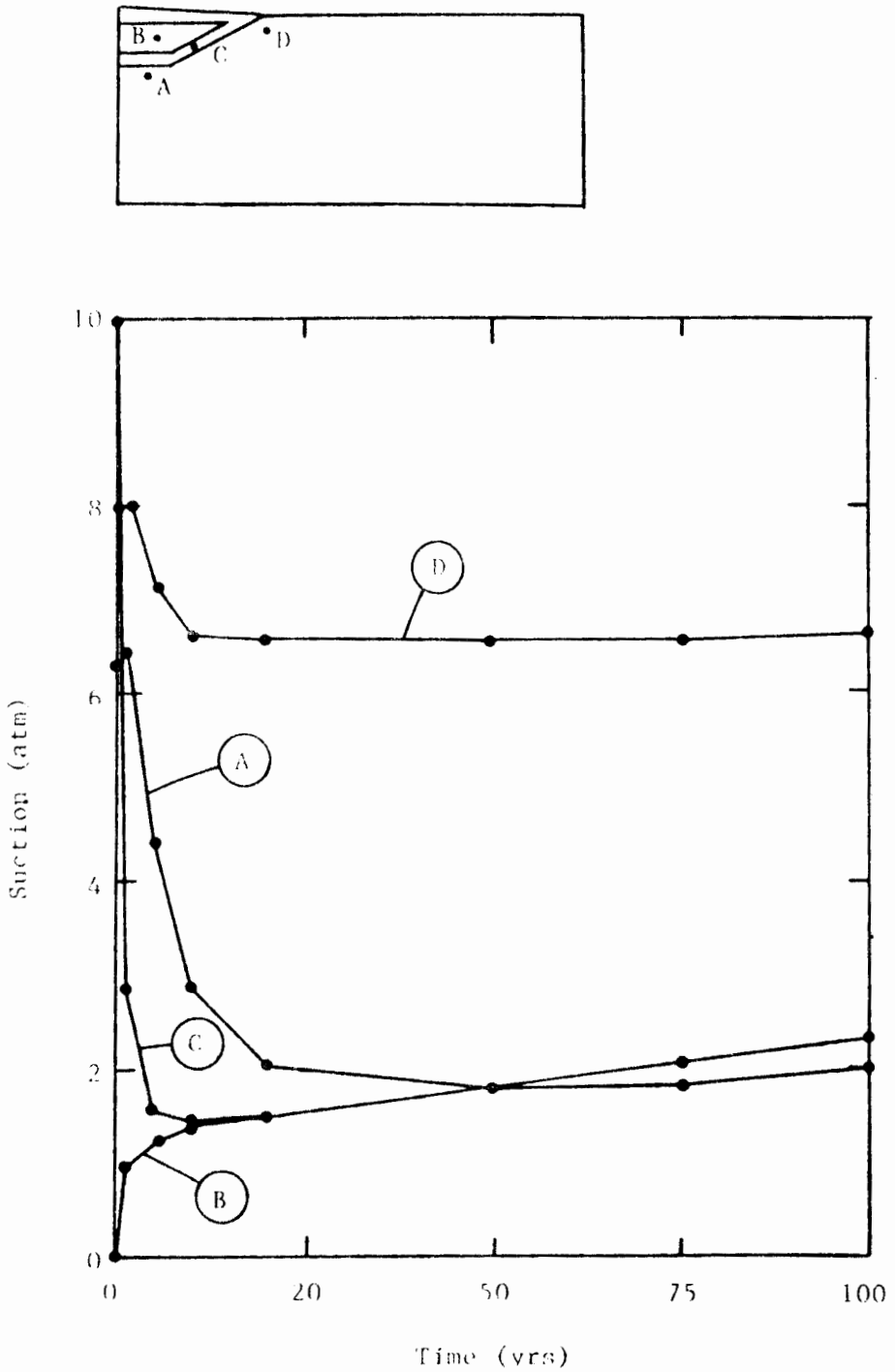


Fig. 4.8-- Variation of Suction with Time at Several Points.

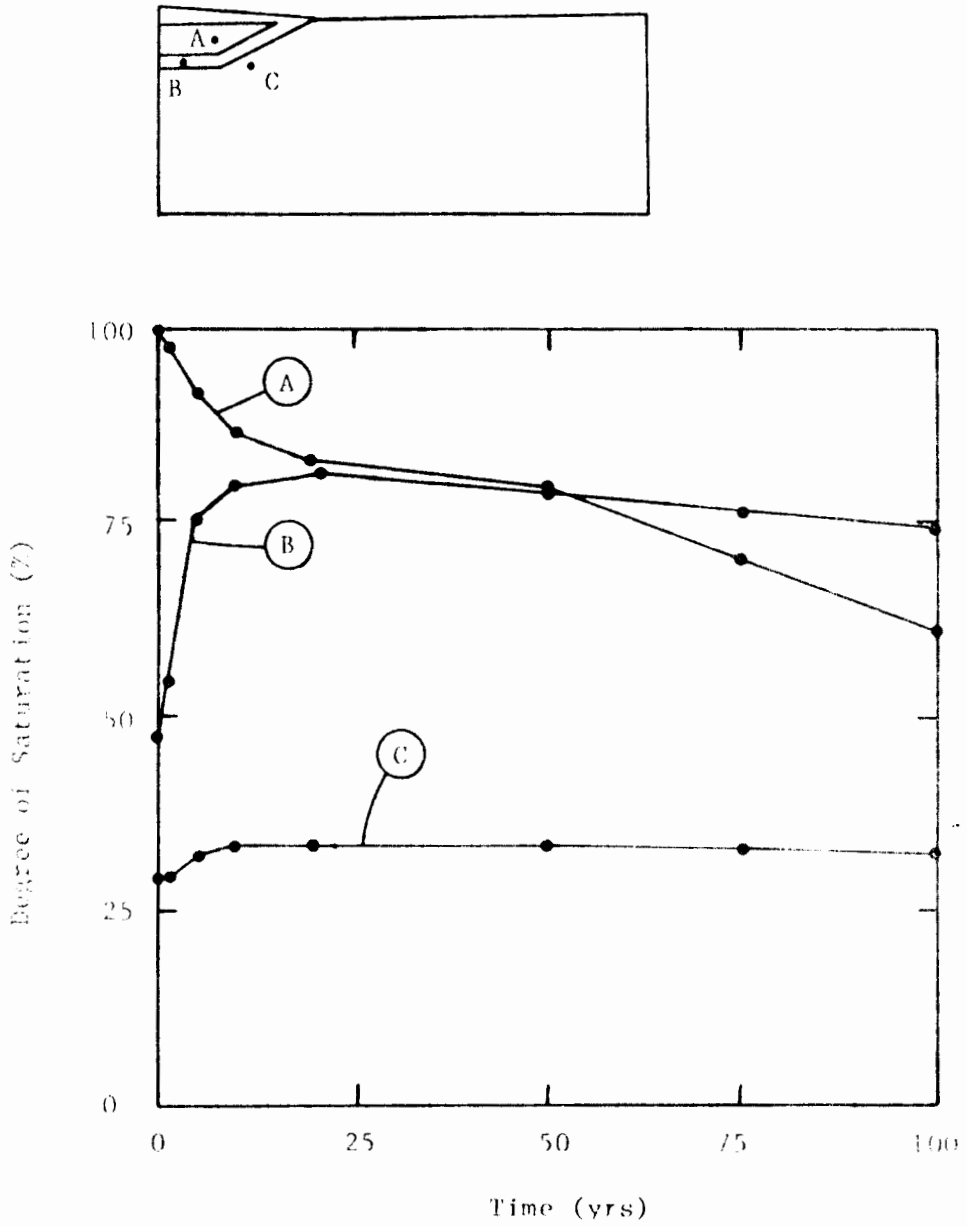


Fig. 4.9-- Variation of Degree of Saturation with Respect to Time at Several Points.

with time in the waste (point A), rises with time in the liner (point B) at first but then gradually drops with time later, and is relatively unaffected several feet beyond the limits of the disposal pit (point C).

A clearer picture of the extent of pollution can be obtained by examining the movement of imaginary tracers. Ten tracers were used and were located initially along the base of the pit, between the waste and liner (Fig. 4.10). The positions at the output times are plotted in Fig. 4.11. Note that the pollutants travel further in the horizontal direction than in the vertical; this is because the saturated conductivity of the liner is assumed to be 10 times larger for horizontal flow than for vertical flow (Table 4.1). The maximum downward movement of pollutants is only about 5 feet below the base of the liner. From these findings, there appears to be little, if any, danger that pollutants would reach the water table within 100 yrs. However, pollutants might be concentrated at the ground surface, near the edge of the pit, as a result of water leaving the pits, flowing laterally and upwards, evaporating, and leaving a residue behind.

4.5.4 Concluding Remarks

The example problem presented here was analyzed to indicate the nature of the results that can be obtained with the computer program, and to document the cost of a typical run. Because of the inevitable uncertainties in soil properties, the user must bear in mind that the results of the program are not exact. The program is best used to show general trends in groundwater pollution and to focus attention on the critical problems. In this example, the critical problem does not seem to be downward seepage of pollutants from the waste disposal pit and into the groundwater supply. A more important problem is concentration of pollutants at the ground surface around the edges of the disposal pit.

Fig. 4.10 -- Initial Positions of Imaginary Tracers.

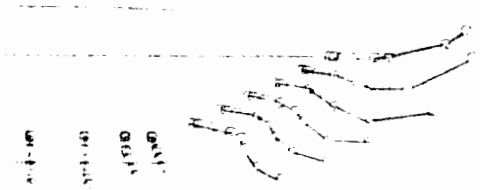


Fig. 4.11 -- Position of Tracers at Calculation Times of 0, .1, 1, 5, 10, 20, 50, and 100 yrs.

5.0

PRETREATMENT OF RADIOACTIVE WASTE

5.1 Introduction

As the nuclear industry continues to develop the attendant volume of generated nuclear waste continues to grow. The practice of disposing of non-high level (NHL) radioactive wastes in shallow land burial sites is being questioned politically and professionally in the areas of safety and volume capacity. The available burial sites cannot possibly contain the amounts of waste being generated unless the wastes are efficiently reduced in volume prior to disposal, and the danger of transporting radioactive material throughout the environment can only be eliminated by carefully controlling, containing, and treating these wastes. This chapter contains a summary of the volume reduction and fixation methods currently used and those being developed for the future. These methods are discussed in detail by Sheppard (1979).

At present, approximately 4 million cubic feet of solid, NHL radioactive wastes are being buried annually at sites around the country and the volume is expected to increase more than ten times by the year 2000. The forms of the wastes generated range from liquid to solid and from combustible to noncombustible. The wastes consist primarily of short-lived beta-gamma emitters, long-lived alpha emitters, or combinations of both. They do not generally yield sufficient radioactive decay energy to pose heat dissipation problems. Sources of NHL wastes include nuclear reactors, nuclear fuel processing and reprocessing facilities, industry, hospitals and research facilities.

Management of radioactive waste is an evolving process. The term "management" has been used to refer to many different aspects of waste handling and treatment, but in its broadest sense it encompasses everything done to the waste from its generation to its disposal. Management can generally be divided into categories of pretreatment, volume reduction, fixation, transportation, and disposal.

5.2 Pretreatment

Pretreatment steps taken during the handling of solid waste include sorting, shredding, and decontamination.

5.2.1 Sorting

Sorting of the waste consists of segregating the waste into groups requiring similar treatment. The sorting method may be by hand or may involve the use of sophisticated equipment which separates waste by radioactive levels or characteristics. For example, systems using air or inert gases can separate combustible from incombustible material based on density differences.

5.2.2 Shredding

Shredding usually applies only to combustible wastes. Shredding itself is a type of volume reduction. However, it usually precedes pneumatic sorting, incineration, or incorporation into a solidification medium.

5.2.3 Decontamination

Decontamination is normally accomplished only on incombustible solid waste. Discarded equipment must frequently receive decontamination treatment. The treatment is normally restricted to reducing the radioactivity to levels which allow handling of the waste.

5.3 Volume Reduction Methods

Limited burial site volume and the difficulty of obtaining new burial sites makes volume reduction essential. The volume of waste being generated may possibly be a more serious problem then the radioactivity itself. This section discusses volume reduction method for gaseous, liquid, and solid wastes.

5.3.1 Treatment of Gaseous Wastes

The type of treatment given to a particular gaseous waste depends on the chemistry and volume of the waste, and the type of radionuclides and level of radioactivity in the waste. The waste may be of such composition that simple dilution with controlled release from a stack is adequate. The waste may also be of such composition that a series of treatment steps are required, such as filtering and scrubbing. Types of filters used to remove entrained particulate matter include packed fiber filters, high efficiency particulate air (HEPA) filters, sand bed filters, and fabric filters. Gaseous radionuclides are removed in scrubbers or in adsorption, absorption, or condensation devices. Virtually all of the radioactivity removed is either in a solid or a liquid form.

5.3.2 Treatment of Liquid Wastes

There are several factors which influence the most effective and economical treatment of liquid waste. Some of these factors include the viscosity, chemical composition, and radionuclide make-up of the waste. The final desired form of the waste is also a factor in determining what treatment method is utilized. If the concentrated radioactive waste is to be mixed with a dry solidification agent, then it would probably have a final presolidified liquid form. If the waste is to be put into a glass or ceramic matrix, its final treatment form will probably be a powder or a dry granular particle.

Various methods of treating liquid radioactive waste include:

• Evaporation. The evaporation process consists mainly of transferring heat to the waste solution and separating the liquid and vapor phases. Because of its high operating cost, evaporators are most often used for solutions containing high concentrations of dissolved solids or solutions which are not treatable by low temperature operations such as precipitation filtration and ion exchange.

• Drying. Drying goes one step beyond the evaporation process in that a dry solid is produced by adding heat until the liquid fraction of the waste is removed.

• Calcination. Calcination goes beyond evaporation and drying of liquid wastes. High temperatures dry and thermally decompose the waste to form oxides.

• Incineration. Liquid wastes which are combustible may be disposed of by incineration methods. Because the wastes are in liquid form, sufficient heat must be supplied above the ignition temperature to vaporize the waste.

• Filtration. Filtration is a process where in liquid waste is filtered through some porous media to remove particles. The porous media may be sand, crushed anthracite coal, diatomaceous earth, perlite, or activated carbon.

• Centrifugation. Centrifugal force is used to separate particles from liquids.

• Ion Exchange. The ion exchange process involves the removal of radioactive material from wastewater by the exchange of ions between the waste and solid ion resin bed. Typically an ion exchange consists of a bed composed of both cation and ion resins. Ion exchange is frequently preceded by coagulation, sedimentation or filtration in order to prevent plugging of the ion exchange bed.

• Membrane. In the reverse osmosis method the waste solution is pressurized above the osmotic pressure and is brought into contact with a semipermeable membrane. The solute passes through the membrane in the direction of the lower osmotic pressure, so that the larger molecules of waste remain on the side of the membrane with the higher pressures.

• Flocculation/Precipitation. Basically, the process of flocculation involves adding chemicals to a waste solution to form a floc which adsorbs contaminants and then precipitates out of solution. The radioactivity may be removed by direct precipitation, by adsorption on floc, or by entrainment in precipitates. The resultant sludge is then further treated and immobilized.

5.3.3 Treatment of Solid Waste

The final form of treated solid waste may range from compressed waste with the same chemical and physical makeup of the original waste to a treated waste which is altered chemically and/or physically from the original waste.

The volume of solid radioactive waste continues to grow yearly. Volume-reduction techniques such as incineration, and the sorting of nonradioactive waste from radioactive waste are viable methods for coping with the large volumes of waste being generated. Volumes of solid waste from commercial sources increased from 1,900 m^3 in 1962 to 57,000 m^3 in 1975. The projected volume of waste by the year 2000 is 240,000 m^3.

Various methods for treating solid waste include:

• Compaction. The simplest and most widely used volume reduction technique is compaction. It is estimated that 50% or more of all solid waste generated is compactable. Estimated factors of volume reduction range from 2 to 10.

• Molten Salt Combustion. This modification of the incineration process uses molten salt to oxidize combustible material. The salts serve as a heat transfer medium, a trap for particulate matter, and a neutralizing agent for acid gases.

• Acid Digestion. Combustible waste is shredded and added to a solution of sulfuric acid at 250° C. Sulfuric acid serves to carbonize the waste. When nitric acid and oxygen is added, the waste is oxidized to carbon dioxide and water.

• Decontamination. Decontamination of radioactive equipment is commonly practiced with the intention of reducing radioactivity to levels which will permit maintenance on or disassembly of the equipment. The liquids normally used in decontamination are highly acidic or highly basic.

• Melting. This method of treatment is used almost exclusively on contaminated metal waste. The metallic wastes are melted and cast into ingots for storage with volume reduction ratios ranging from 5/1 to 15/1.

• Incineration. Incineration not only reduces waste volumes by factors ranging from 20 to 50 but also renders the waste chemically inert. Combustion is becoming the most effective means of treating solid waste. An estimate of costs for incinerating solid waste is shown in Table 5.1.

5.3.4 Discussion

In order to establish whether volume reduction should be required, the areas of waste handling, transportation, and disposal must be addressed. Some advantages of volume reduction in these areas are obvious. Personnel exposure is less during packaging, loading, and unloading operations because less waste is handled. Transportation is safer because fewer trips must be made and therefore the probability that accidents will occur is decreased. And finally,

TABLE 5.1

28-WEEK RADWASTE INCINERATOR OPERATION SUMMARY*

$ 5,200	Filter costs
2,700	Labor costs
18,000	Total cost of incinerator and expense on it to date including $6,000 test expense
$25,900	

Cost of disposition to previous source	$108,000
Actual cost of disposition including incinerator	25,900
Net Savings through August 22, 1960	$ 82,100

*1960 dollars.

SOURCE: Chandler, R. J., and Thorburn, R. C., "An Incinerator for Uranium Contaminated Wastes," General Electric, Report No. GEAP-3542, p. 8 (1960).

reduced volume means extended time to fill available disposal sites.

The economic aspects of volume reduction are not obvious, however, except for burial costs which are related to waste volume and are estimated to be between \$20 to \$200/m^3 (Takamura, 1979). The cost effectiveness of volume reduction in regard to handling and transportation depends on such factors as level of radioactivity level, weight of waste, volume of waste, and distance to burial site.

Reducing waste volumes concentrates radioactivity which leads to higher radioactivity levels, and may require more shielding and higher container costs. These increased costs tend to offset cost decreases in other areas. Waste container requirements, limitations on package contents, and packaging and handling procedures are contained in Title 49 and Title 10 of the Code of Federal Regulations.

5.4 Fixation Methods

5.4.1 Current Methods

Fixation of the waste immobilizes the radionuclides in such a manner that contamination of the environment is prevented or greatly minimized. The solidification of the waste also facilitates handling, transportation, and disposal operations.

One of the tradeoffs that one must face when contemplating fixation is whether the increase in volume, that is a result of fixation methods, is acceptable. Generally the waste volume increase is acceptable because there is a net decrease in the original volume when the waste is pretreated.

Most of the fixation methods currently used apply to treatment of liquids, sludges, ion exchange resins, and other water-saturated wastes. Table 5.2 compares the various techniques.

TABLE 5.2

COMPARISON OF IMMOBILIZATION TECHNOLOGIES

Comparison Factor	Absorbent	Salt Cake	Urea-Formaldehyde	Portland Cement[a]	Bitumen
Shelf life of immobilizing agent:	Long	NA[b]	Short (months)	Long	Long
Mix fluidity	Fair	NA	Good	Poor	Fair
Mixer cleanability	Good	NA	Good	Poor	Fair
Chemical tolerances					
Boric acid Sol'n	--	--	Good	Poor	Good
Na_2SO_4 Sol'n	--	Good	Reduced efficiency	Fair	Good
$NaNO_3$ Sol'n	--	Good	Fair	Good	Not recommended
Alkaline Sol'n	--	--	Reduced efficiency	Good	Good
Laundry det. Sol'n	Good	--	Poor	Poor	Fair
Organic liquids	Good	NA	Poor	Poor	Fair
Ion exch. resins	NA	NA	Good	Fair	Fair
Sludges	NA	NA	Good, may require pH adjustment	Good	Good; hazardous with MnO_2 sludge
Volumetric Efficiency[c]	Low(0.3-0.5)	High(4-8)	Moderate(0.6-1.0)	Low (0.5).	High(>2)
Product Form	Moist granules or paste	Monolithic cake	Monolith	Monolith	Monolith
Product density, kg/m^3	400-1000	2000-2500	1000-1300	1500-2000	1000-1300
Water binding strength	Poor	Water evap. during preparations	Fair	Good	Water evap. during preparations
Residual free water	Occasionally	Occasionally	Occasionally	Seldom	Never
Mechanical strength	Nil	Low	Fair	Good	Good
Product stability	Poor	Fair	Fair (loses water and strength in open system)	Very good	Good
Combustibility	Depends on liquid absorbed	No	Yes		Yes
Freeze/thaw resistance	NA	NA	Poor	Fair to good	Good
Leach resistance	Very low	Low	Moderate	Moderate to high	High

[a]Without additives
[b]NA = not applicable
[c]Ratio of the as-generated waste volume to the solidified waste volume

SOURCE: U.S. Energy Research and Development Administration, "Alternatives for Managing Wastes from Reactors and Post-Fission Operations in the LWR Fuel Cycle," Vol. 2 of 5, U.S. ERDA Report No. ERDA-76-43, p. 12.4 (1976).

• Absorption. Granular or powdered material has been used to immobilize liquid waste by holding waste molecules in its pores. The bond between the absorbent and the waste molecule is not a chemical bond, and consequently the entrapped radioactive liquid can be displaced by the addition of water. Due to their high leachability, absorbents are used mostly for organic and inorganic liquids that are not easily treated by any other method.

• Salt matrix. Waste solids are incorporated into a salt matrix. Salt cakes are highly leachable, but because solified salts are neutral, they pose minimal corrosion problems and can therefore be stored in metal containers.

• Cement. Hydraulic cement has been used extensively for many years as a means of fixing radioactive wastes. The curing process of the cement mixture is not entirely understood but involves the reaction of waste with the cement in such a manner that the oxides crystallize. The type and amount of radioactive waste added to the cement, e.g., whether the added waste is liquid or solid, the total salt content of the waste, and the pH of the waste, affect the properties of the hardened product.

• Bitumen. Bitumen or asphalt has been used extensively in foreign countries. The bitumenization process consists of mixing the waste sludge or solids with emulsified asphalt or molten-base asphalt. The temperature is then raised to evaporate the water. About 99% of the water is removed which results in a volume reduction five times as great as that achieved in cement processes.

• Polymerics. Using polymeric fixation agents to contain radioactive waste is a new process compared with the use of cement and bitumen. Several types of polymers have been used, including paraffin, polyethylene, and urea formaldehyde.

5.4.2 Methods under Development

The following techniques are high-priced and were developed for use on high-level wastes. It is unlikely these methods would be used in the pre-treatment of low-level waste and are included only for completeness.

Clay. Once a liquid waste has been treated and reduced to a salt, it becomes possible to incorporate the salt in a clay such as aluminosilicate using a hydrothermal process. Powdered kaolin or bentonite clay may be used at temperatures ranging from 30° to $100^{\circ}C$ to form a cancrinite mineral which contains the waste salt as part of its crystalline structure.

Glass. Glass is chemically durable and can contain a wide variety of wastes. Glass research has been ongoing for nearly 25 years with most of the emphasis placed on containing high-level wastes. Fixation of non-high level waste is a much simpler process because most of this waste consists of sodium salts instead of fission products.

Pelletization. Pelletization may be used with waste salts, calcines, and incinerator ash, and consists basically of subjecting the waste to a pressing process. The waste may be hot or cool during processing. The product is generally less soluble and leachable than loose waste. If a ceramic matrix is added, the product will be highly leach resistant. It is also possible to make the surface of the pellet inert with ceramic, metal, or epoxy coatings.

Coating. This method is an extension of encapsulation and pelletization technology and involves the use of solidification agents to coat the waste and increase its leach resistance.

Metal Matrix. Metal matrices are normally used in waste fixation to provide strength and thermal conductivity.

• Supercalcination. This process involves mixing additives with the waste sludge so that after calcination and heat treatment, a crystalline ceramic waste is formed. The ceramic has a higher leach resistance than unmodified calcine.

• Sintering. Sintering involves subjecting waste to high pressures in order to form a concentrated mass.

• Ceramics. Incorporation of radioactive waste in a ceramic matrix is very similar to glass processes. The major difference between glass and ceramic is the type of additives added to the waste and the levels of temperature utilized.

5.5 Transportation

There are several advantages in treating the waste prior to burial. One advantage is that treated waste is easier to handle and safer to transport. In addition, the number of containers and transportation costs are reduced for solidifed waste.

Transportation costs may be applied on a weight or volume basis. Volume reduction is less attractive if a weight basis is used because the reduction in weight is generally much less than the reduction in volume. Depending on the fixation method used, the weight reduction after volume reduction may be entirely offset by the weight of the fixation agent added. Table 5.3 contains an estimate of transportation costs based on weight of waste processed.

5.6 Disposal

Waste disposal costs increase as the volume of waste increases. As a result, the volume reduction methods discussed in this chapter will be come more cost effective in the future when the volume of wastes increase and the available disposal spaces decrease.

TABLE 5.3

UNIT COST ESTIMATE FOR TRANSPORT OF NHL WASTE IN TERMS OF COST PER KG HM PROCESSED

Waste Immobilization Process	Surface Radiation Rate R/hr	No. of Waste Containers/MTHM				Unit Cost, $/kg HM				Total
		55-Drums	80-gal Drums	Boxes	In. 30-in dia Canisters	55-gal Drums	80-gal Drums	Boxes	In. dia Canisters	
Incineration with cementation of ash and wet wastes (reference process)	<0.2	2.432	-	0.035	-	0.15	-	0.03	-	0.18
	0.2-1	0.976	-	-	0.034	0.11	-	-	0.43	0.54
	1.0-10	0.983	-	-	0.002	0.27	-	-	0.03	0.30
	>10	2.129	-	-	-	1.43	-	-	-	1.43
Total						1.96		0.03	0.46	2.45
Incineration with bituminization of ash and wet wastes	<0.2	1.338	-	0.035	-	0.08	-	0.03	-	0.11
	0.2-1	1.728	-	-	0.034	0.19	-	-	0.43	0.62
	1.0-10	0.983	-	-	0.002	0.27	-	-	0.03	0.30
	>10	0.943	-	-	-	0.63	-	-	-	0.63
Total						1.17		0.03	0.46	1.66
Minimum treatment with cementation of wet wastes	<0.2	7.317	-	0.035	-	0.44	-	0.03	-	0.47
	0.2-1	1.336	-	-	0.034	0.15	-	-	0.43	0.58
	1.0-10	4.223	-	-	0.002	1.14	-	-	0.03	1.17
	>10	1.551	1.450	-	-	1.04	0.97	-	0	2.01
Total						2.77	0.97	0.03	0.46	4.23
Minimum treatment with bitumination of wet wastes	<0.2	7.338	-	0.035	-	0.44	-	0.03	-	0.47
	0.2-1	1.666	-	-	0.034	0.18	-	--	0.43	0.61
	1.0-10	4.223	-	-	0.002	1.14	-	-	0.03	1.17
	>10	0.615	1.450	-	-	0.41	0.97	-	-	1.38
Total						2.17	0.97	0.03	0.46	3.63

SOURCE: U.S. Department of Energy, "Technology for Commercial Radioactive Waste Management," Vol. 4 of 5, Report No. DOE/ET-0028, p. 6.6.11 (1979).

6.0

BIOLOGICAL MOBILIZATION OF CONTAMINANTS

In the past, off-site migration of contaminants at shallow land burial sites has been a problem. The influence of organic matter and biological activity on the mobilization of radioactive waste is discussed by Herrera (1979) and summarized below.

The analysis is based on the following assumptions:

1. The wastes are not pretreated to remove organic matter;
2. There will be enough moisture present in the waste to sustain bacterial activity;
3. There will be enough moisture in the soil to sustain plant growth; and
4. The effects of any barriers around the site, excluding in situ soils, are disregarded.

6.1 Mobilization by Bacterial Activity

One of the most important biological mechanisms in mobilizing contaminants is bacterial activity. Bacterial activity tends to lower the pH which, in turn, increases the solubility of radionuclides. A low pH also decreases the sorption capability of soils.

The sequence of events in the bacterial decomposition of organic matter in a burial site is illustrated in Fig. 6.1. The basic phases are aerobic and anaerobic decomposition followed by methane fermentation. After anaerobic fermentation has been established, the concentration of volatile acids in solution increases. When volatile acids attain their maximum concentration, the solubility of leachable ions such as Strontium-90, Cesium-137, and Cerium-144 also peaks so that mobilization is at its maximum. As the methane

Event	Effects
Burial of wastes	
↓	
Water intrusion (ground water or percolation from surface water)	Initiation of bacterial activity.
↓	
Aerobic decomposition (very limited)	Mobilization of C-14 in bacterially metabolized CO_2 plus some mobilization of radionuclides with the leachate
↓	
Anaerobic decomposition, formation of volatile acids, and lowering of pH (acid fermentation)	Mobilization of C-14 in the form of CO_2 and enhanced mobilization of soluble radioisotopes with the leachate.
↓	
Methane fermentation	Mobilization of C-14 and tritium as CO_2 and CH_4 with gradual increase in pH and decreased concentration of soluble nuclides in the leachate

Fig. 6.1 Sequence of Events in the Bacterial Decomposition of Organic Matter in a Burial Site

bacteria utilize the volatile acids, the pH rises and leachable ions precipitate out of solution. At this point, mobilization of contaminants such as C-14 and tritium may increase through gaseous releases.

6.2 Mobilization by Coupling with Organic Compounds

Chelating agents may provide a key role in allowing contaminants to migrate freely through the soil. A chelating agent consists of a ring of atoms in which a metal ion is held in a coordination complex. Once a bond has formed between the metal ion and chelating agent, the ring is complete and the compound may travel through the soil without being attenuated.

Chelating agents may be present in the waste at the time of burial or they may be formed during the decomposition of organic matter. One chelating agent frequently found in the waste has been EDTA. Other chelating agents of importance are NTA and TTHA. NTA forms stable complexes with most metals. TTHA has an affinity for thorium, zirconium, indium and gallium. A wide range of chelating agents such as fluvic acids, organic acids, amino acids, and phenolic compounds are formed in soils during the decomposition of organic matter, root exhudation, and microbial synthesis by rhyosphere microorganisms.

A lowering of the pH by bacterial activity may either increase or decrease the activity of the chelating agents. It may also decrease the solubility of some organic compounds which would decrease the mobility of the nuclides coupled with these compounds.

6.3 Mobilization by Plants and Burrowing Organisms

6.3.1 Plants

In general, uptake of radionuclides by plants is inversely related to pH (higher solubilities occur at lower pHs) and to organic matter content (organic matter competes with the nuclides). Sorption is thus greatest in

acidic soils with low organic content and lowest in calcareous soils.

The nuclides most highly concentrated by plants are alkaline metals, halides, and essential nutrients of vegetables. Soluble minerals and those minerals that are weakly adsorbed by soil tend to be concentrated in the greatest amounts.

6.3.2 Burrowing Organisms

Mobilization of radionuclides by burrowing animals may occur by contaminated soil being brought to the surface or by contaminants being transported in the claws, fur, etc. of the animal. Another possible means of transport may occur if the waste is ingested and later deposited in the form of body wastes. Possible effects of various burrowing organisms in the mobilization of waste is shown in Table 6.1.

6.4 Recommendations

There are several possibilities for minimizing biological release of contaminants from disposal pits:

1. Build barriers around the site to prevent the infiltration of water and the intrusion of plant roots and burrowing organisms. This alternative is ideal in theory but it is doubtful that present technology can provide the materials and methods required to accomplish this feat for the life of the site.

2. Locate the site in an area of low precipitation, low population density and deep ground water table. These areas are concentrated in the western and southwestern portions of the U.S., whereas the bulk of the wastes are produced in the eastern and midwestern regions. The problems of safe transportation and handling, as well as legal problems with the states where these areas are located, may well prove more difficult to resolve than

TABLE 6.1

Effects of Various Burrowing Organisms in the Mobilization of Wastes

Organism	Type of Mobilization
Reptiles	Little or none due to the shallow depth of the burrow
Mammals	
Moles	Little or no solid mobilization due to the shallow depth of the burrows. Increased mobilization of gaseous radionuclides in the top layer of soil through the extensive tunnel network
Badgers	Mobilization of solid radionuclides through digging. Mobilization of radioactively contaminated materials which may be appropriate for bedding. Intake of radioactively contaminated plants to the burrow for bedding. Tunnels can provide a pathway for gaseous radionuclides and for the infiltration of water
Marmots	Mobilization of solid radionuclides through digging. Intake of radioactively contaminated plants for padding. Tunnels provide escape pathways for gaseous radionuclides and for the infiltration of water
Small Animal Species	
Earthworms	Transport of solid radionuclides out of the site through digging and by deposition of ingested soil in the surface. Increased porosity of the soil cap through the tunnels can provide a pathway for

TABLE 6.1 Cont.

Organism	Type of Mobilization
Small Animal Species	
Earthworms	gaseous radionuclides and for the infiltration of water. Intake of organic matter from the surface can provide fuel for bacterial activity. Spread of radionuclides by worm eating birds can also take place. Introduction of chelating agents formed from litter decomposition in the top soil
Ants	Removal of solid radionuclides through digging. The intricate tunnel net can provide pathways for gaseous radionuclide escape and water infiltration. Introduction of chelating agents formed during litter decomposition in the top soil

considering an alternate method of disposal.

3. Promote rapid biological activity and radionuclide leaching such that most leachable ions are leached out of the site and decomposition of organic matter is completed in a short period of time. Treat the leachate by ion exchange or other suitable methods and collect the gaseous radionuclides. The collected materials can then by stabilized and stored in the site. Uptake by plants and burrowing animals as well as complexing with organic matter will be minimal since the treated wastes will be essentially free of radionuclides. With the technology available at present this is perhaps the best alternative.

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