NUREG/CR-4897
BNL-NUREG-52066

LOW-LEVEL WASTE SOURCE TERM EVALUATION: REVIEW OF PUBLISHED MODELING AND EXPERIMENTAL WORK, AND PRESENTATION OF LOW-LEVEL WASTE SOURCE TERM MODELING FRAMEWORK AND PRELIMINARY MODEL DEVELOPMENT

TOPICAL REPORT

T. Sullivan and C.R. Kempf

February 1987

NUCLEAR WASTE MANAGEMENT DIVISION
DEPARTMENT OF NUCLEAR ENERGY, BROOKHAVEN NATIONAL LABORATORY
UPTON, NEW YORK 11973

Prepared for Office of Nuclear Regulatory Research
U.S. Nuclear Regulatory Commission
Washington, D.C. 20555
Under Contract No. DE-AC02-76CH00016

NUREG/CR-4897
BNL-NUREG-52066
RW

LOW-LEVEL WASTE SOURCE TERM EVALUATION: REVIEW OF PUBLISHED MODELING AND EXPERIMENTAL WORK, AND PRESENTATION OF LOW-LEVEL WASTE SOURCE TERM MODELING FRAMEWORK AND PRELIMINARY MODEL DEVELOPMENT

TOPICAL REPORT

T. Sullivan and C.R. Kempf

February 1987

NUCLEAR WASTE MANAGEMENT DIVISION
DEPARTMENT OF NUCLEAR ENERGY
BROOKHAVEN NATIONAL LABORATORY
UPTON, LONG ISLAND, NY 11973

Prepared for
OFFICE OF NUCLEAR REGULATORY RESEARCH
UNITED STATES NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555
UNDER CONTRACT NO. DE-AC02-76CH00016
NRC FIN A3276

Available from
Superintendent of Documents
U.S. Government Printing Office
P.O. Box 37082
Washington, DC 20013-7982
and
National Technical Information Service
Springfield, Virginia 22161

ABSTRACT

This report contains information on the efforts performed to date on the Low-Level Waste Source Term Evaluation Project, the objective of which is development of a model to predict radionuclide release rates from a low-level waste disposal unit. The approach for model development has been based on a compartmentalized scheme focused on the four major processes of water flow, container degradation, waste leaching and waste radionuclide transport to the trench boundaries. This stage of the project is focused primarily on modeling release rates from shallow land burial as currently practiced. Research efforts to this point include characterization work (of burial trenches themselves, of soils and structural features, and of waste forms and containers), review of published modeling work, review of several waste package performance system models, and development of original container degradation and waste leaching models. Characterization of the wastes, containers, and of the site (trench soils and structure) has been based on the premise that NRC guidance has been put into effect. Quantitative prediction of the water flow in the trench is a major part of this program. The water flow equations are generally formulated to yield the hydraulic potential in the porous medium as a function of space and time. The solution of the water flow equations provides a method to obtain the average velocity with which the water moves at each spatial location in the disposal unit. This velocity is used in the contaminant transport equation. Inclusion of waste package container degradation and waste leaching leads to the radionuclide release amounts which must be coupled with the water flow and contaminant transport models to complete the scheme.

CONTENTS

CONTENTS (continued)

LIST OF FIGURES

LIST OF TABLES

ACKNOWLEDGMENTS

The authors extend their appreciation to A. Spira for her efforts in the preparation of the report manuscript.

EXECUTIVE SUMMARY

This is the first topical report for the Low-Level Waste Source Term Evaluation Project, which was initiated in June, 1985 with the purpose of providing an estimation of the rates of radionuclide release from a low-level waste disposal unit.

Before the issuance of the NRC Final Rule on Licensing Requirements for Land Disposal of Radioactive Waste (10 CFR 61) in 1982, low-level wastes were routinely disposed of in shallow-land burial sites in unsegregated, unconsolidated, as well as poor-integrity consolidated conditions. Although burial trenches were backfilled with soil, and caps were installed over the trenches, subsequent compaction of the wastes and backfill often led to instances of trench subsidence and enhanced water accumulation around the waste. Concerns about the potential for accelerated leaching of radionuclides from the waste, and their eventual transport to the accessible environment, prompted the development of more stringent site and package criteria for shallow land burial. These are specified in Rule 10 CFR 61, the NRC Technical Position on Waste Form, and the NRC Technical Position on Site Suitability, Selection and Characterization. Historically, the contents of many existing shallow land burial trenches at both closed and operational sites in the United States have been poorly characterized and radionuclide release rates (source terms) have been essentially unknown.

The approach to radionuclide release rate estimation involves development of a model; this has been designed as a compartmentalized scheme involving the four major processes of water flow, container degradation, waste leaching and waste radionuclide transport to the trench boundaries.

This stage of the project is focused primarily on modeling release rates from shallow land burial as currently practiced. Efforts are being made and will continue to keep the methodology general enough to allow the radionuclide release rates from alternative disposal approaches to be evaluated with minor modifications to the solution techniques.

Research efforts to this point include characterization work (of burial trenches themselves, of soils and structural features, and of waste forms and containers), review of published modeling work (the vast majority of which represents water flow codes and transport codes with assumed source term values or functions) review of several waste package performance system models used to evaluate the expected dose to the population resulting from low-level waste disposal; and development of original container degradation and waste leaching models.

Characterization of the wastes, containers, and of the site (trench soils and structure) has been based on the premise that NRC guidance has been put into effect. This means that wastes can be expected to consist of two main types: non-stabilized, heterogeneous, relatively low activity, Class A; and stabilized (through solidification to a monolithic waste form or through placement in a high integrity container), generally physically more homogeneous, relatively higher activity, Class B and C. Within each of these

two categories of waste there can exist tremendous variety in waste radionuclide species, their chemical forms and their physical form as well as variety in the containers used. Performance of the Class B and C waste packages may be expected to coincide, as a minimum, with the recommendations given in the NRC technical position. Trench design and waste emplacement and backfilling practices should lead to minimal worker exposure and maximal trench stability. The specifics of waste classification, minimum waste package requirements and burial site practices have been reviewed and are summarized in this report. It is understood that information on waste inventory and chemical and physical characteristics forms a primary segment in the foundation on which any source term modeling would rest.

To minimize any duplication of effort in this modeling program an active and ongoing review of published models pertinent to water flow, contaminant transport, and radioactive waste package performance has been carried out.

A number of unsaturated water flow codes which may be adapted to fill the requirements of the source term project have been identified. From these codes, UNSAT2 and FEMWATER are currently under examination for use in predicting water flow in the disposal unit. Both codes are considered to be state-of-the-art and have been used by NRC staff. Similarly, a number of contaminant transport codes have been identified. FEMWASTE, the companion code to FEMWATER, is being reviewed for potential use in the source term project.

Highlights of BNL's waste package radionuclide release rate modeling effort for the Sheffield burial site have been summarized for H-3, C-14, Cs-137, Sr-90 and Co-60 containing wastes. This modeling included consideration of idealized diffusion, dissolution, permeation and radiolysis mechanisms as well as incorporation of empirical findings applicable to releases from polymer materials, metal hydrides, and concrete blocks.

Experimental work relevant to predicting radionuclide release from a disposal unit was reviewed. These experiments tend to focus on developing an understanding of waste form leaching and radionuclide transport in an unsaturated soil. Water flow is studied in relation to transport.

The three major experiment categories are: 1) leaching experiments which provide information on the interaction of the waste form and solution; 2) column tests in which a tracer is injected at the top of the column, both water flow and contaminant transport in a porous medium are examined; and 3) lysimeter tests which involve a waste form surrounded by a porous soil, information on water flow, leaching, and transport is obtained. The data from these experiments should be useful in obtaining parameters for leaching and contaminant transport models. The results of these experiments (particularly the lysimeter tests) should be useful in validating the models.

Container performance has received less attention than leaching and contaminant transport in relation to low-level waste disposal. However, for steels and concretes, there is a large data base of information useful to source term modeling on their properties and performance. Much less data is available for high density polyethylene.

The four main model compartments of water flow, container degradation, waste leaching and waste radionuclide transport are each comprised of multiple subparts. Water flow will occur through the trench cap, then through backfill in and around waste packages and then, (once container degradation and waste leaching have occurred), laden with radionuclides, it will flow to the trench boundaries.

The water flow equations are generally formulated to yield the hydraulic potential in the porous medium as a function of space and time. The key parameters required for solution of this equation are the hydraulic conductivity, which is a function of soil moisture content, and sources or sinks of water. Sources include precipitation and seepage into the disposal unit. Sinks include evaporation, transpiration, and drainage out of the trench.

A model for container degradation and surface species leaching from porous wastes is presented in this report. In short, this model consists of restricted water infiltration through corroded areas in the outer waste container; filling of the pore space volume of the waste with concurrent uniform leaching of radionuclide species residing on pore space surfaces; and then, exit of leachate at a rate driven, at long times, by a steady state approximation in the waste package as a whole (i.e., exiting leachate volumes match incoming water/leachant volumes).

After container breach and waste form leaching occurs, contaminant transport becomes important. The solution of the water flow equations provides a method to obtain the average velocity with which the water moves at each spatial location in the disposal unit. This velocity is used in the contaminant transport equation. Other parameters that are important in the contaminant transport equation are the dispersivity of the contaminants in the soil, and source or sinks for the contaminants. Sources include release from the various waste forms due to leaching, soil/water interactions, and uptake by plants.

1.0 INTRODUCTION AND BACKGROUND

The Nuclear Regulatory Commission (NRC) has the responsibility of regulating and licensing the commercial and nondefense governmental use of source, byproduct, and special nuclear material. This responsibility includes licensing commercial disposal of low-level waste. NRC's responsibility in regulating low-level waste is specified primarily in 10 CFR Part 61, Licensing Requirements for Land Disposal of Radioactive Waste.

In 10 CFR 61 there is a requirement that any near surface disposal site be capable of being characterized, analyzed, and modeled. One intent of this requirement is to ensure that the consequences of radioactive waste disposal can be estimated. Predictive modeling of the release of radionuclides from a proposed burial site based on site characterization will be useful in assessing the suitability of proposed sites for disposal. Monitoring will permit an evaluation of actual site performance.

The objective of this program is to assist NRC in developing the ability to model a disposal site. In particular, a general computer model capable of predicting the quantity and rate of radionuclide release from a disposal trench, i.e., the "source term", is being developed. It involves an estimation of the contents of a "typical" trench and the physical, chemical, and hydrological processes which influence the release of radionuclides to the boundaries of the trench. Specifically, consideration is being given to modeling the rates of water infiltration, container degradation, waste leaching, and radionuclide migration within the trench.

The results of this modeling work should have the following benefits:

a) It will provide the "source term" for geohydrologic calculations which estimate the rate of transport of radionuclides to the accessible environment. From these, a determination may be made whether a site may be safely licensed, operated, closed, and decommissioned.

b) It will allow identification of the important processes and parameters which need to be controlled to minimize the release of activity from a trench and burial site.

c) It will lead to an identification of key data gaps for which critical experiments may be designed and undertaken.

Development of a model to predict the "source term" requires definition of the system to be modeled. The first step in defining the system is to determine the types of waste disposed in a trench and how this waste is emplaced in a trench. This must be followed by characterization of expected water flow quantities and patterns, water-waste package interactions and, then, water flow and radionuclide transport to the trench boundaries. The following sections discuss waste classification, characteristics, and disposal practices. Since disposal practices have changed due to the requirements in

10 CFR 61, the sections of this chapter that are devoted to disposal practices are divided into pre 10 CFR 61 and post 10 CFR 61 practices. It is recognized that future burial sites may not use the trench burial disposal concept; alternative burial techniques are briefly discussed at the end of the post 10 CFR 61 section.

1.1 Summary of 10 CFR Part 61 Low-Level Waste Classification and Waste Package Requirements

1.1.1 Classification

Low-level radioactive waste must meet the requirements specified in 10 CFR Part 61 if it is to be considered acceptable for shallow land burial. The waste must be classified according to the scheme presented in Section 61.55 of the regulation, and also it must conform to the specifications on waste characteristics given in Section 61.56. The guidelines for classifying low-level radioactive wastes (LLW) are based on the concentration and type of radioactive species present. There are three classes: A, B, and C, and these are determined for a particular waste package according to the criteria listed in 10 CFR 61.55 and summarized here.

The first consideration is whether the package contains any of the long-lived radionuclides listed in Table 1 of Section 61.55. Table 1 also gives limiting concentrations for these radionuclides, and these are reproduced here in Table 1.1.1 in which the concentration limits for Class A wastes are explicitly presented in units of curies per cubic meter, as well as in units more amenable to comparison with values encountered on radioactive shipment records (RSRs). If the concentration of a radionuclide exceeds the value given in Table 1.1.1 but does not exceed ten times this value, it is Class C.

If more than one of the radionuclides listed in Table 1 (or, here Table 1.1.1) is present, then the sum of fractions rule is applied. This rule can be represented as follows:

$$SF = \sum_i \frac{RN_i}{RNL_i}$$

where RN_i = radionuclide concentration in the waste package and, RNL_i = concentration limit for that particular radionuclide from Table 1.1.1. As long as SF, the sum of the fractions calculated for the different radionuclides, is less than 1.0, the waste is Class A.

If the waste does not contain any of the long-lived radionuclides listed in Table 1.1.1, then the presence of short-lived radionuclides is considered. In 10 CFR Part 61, the concentration limits for Classes A, B, and C of several radionuclides are listed in Table 2 of Section 61.55. The limits for Class A wastes are reproduced here in Table 1.1.2. If none of the radionuclides listed in Table 1.1.2 is present in the waste, it is Class A. If a combination of the short-lived radionuclides is present, the sum of fractions rule must be applied, and the calculated value of SF must not exceed 1.0.

Table 1.1.1 Concentration limits of long-lived radionuclides for Class A wastes.[a]

Radionuclide	Concentration Limit			
	Ci/m^3	Ci/ft^3	Ci/55 gal drum[b]	Cix17H drum[c]
C-14	0.8	0.023	0.17	0.26
C-14 (IAM)[d]	8.0	0.23	1.7	2.6
Ni-59 (IAM)[d]	22.0	0.62	4.58	7.16
Nb-94 (IAM)[d]	0.02	0.00057	0.0042	0.0065
Tc-99	0.3	0.0085	0.062	0.098
I-129	0.0008	0.00023	0.0017	0.0026
TRU ($t_{1/2}$>5 yr)[e]	10[f]	--	--	--
Pu-241	350[f]	--	--	--
Cm-242	2000[f]	--	--	--

[a]Calculated from values given in Table 1, 10 CFR Part 61.
[b]55-gallon = ≈7.5 ft^3.
[c]x17H drum = 11.5 ft^3.
[d]IAM = in activated metal.
[e]TRU = α-emitting transuranic nuclides (half-life greater than 5 years).
[f]Units are nanocuries per gram.

Table 1.1.2 Concentration limits of short-lived radionuclides for Class A wastes.[a]

Radionuclide	Concentration Limit			
	Ci/m^3	Ci/ft^3	Ci/55 gal drum[b]	Cix17H drum[c]
All with $t^{1/2}$ < 5 y[d]	700	19.8	145.6	227.9
H-3	40	1.13	8.32	13.0
Co-60	700	19.8	145.6	227.9
Ni-63	3.5	0.099	0.728	1.139
Ni-63 (IAM)[e]	35	0.99	7.28	11.39
Sr-90	0.04	0.0011	0.008	0.013
Cs-137	1	0.028	0.208	0.32

[a]From Table 2 in 10 CFR Section 61.55.
[b]55-gallon = ≈7.5 ft^3.
[c]x17H drum = 11.5 ft^3.
[d]i.e., all radionuclides with half-life less than 5 years.
[e]IAM = in activated metal.

Class B wastes may not contain any long-lived radionuclides. If short-lived radionuclides are present in a waste package, the guidelines based on Table 2 of 10 CFR Part 61 must be followed. The radionuclide limits for Class B and Class C wastes are reproduced here in Table 1.1.3. If more than one radionuclide of either type, i.e., short- or long-lived, is present, then the sum of fractions rule must be applied as with the Class A wastes.

If a combination of long- and short-lived radionuclides is present in a waste package, it can be Class A provided the limits in 10 CFR 61 Tables 1 and 2 are not exceeded.

If the Class C limits of either long-lived or short-lived radionuclides are exceeded, the waste is generally considered not acceptable for shallow land burial.

1.1.2 Waste Package Requirements

The minimum waste characteristics requirements for low-level wastes are given in 10 CFR Section 61.56. These requirements deal with the chemical and physical nature of the waste package. Section 61.56 specifies that cardboard and fiberboard boxes cannot be used for packaging wastes. Liquids are required to be solidified or packaged in an amount of absorbent sufficient to absorb twice the volume of liquid. In solid wastes containing liquid, the liquid may not exceed one percent of the volume.

Chemical stability with respect to detonation, explosive decomposition, and explosive reaction with water is also required of the wastes. Generation or containment of toxic gases, vapors or fumes which could be harmful to people is disallowed, as well as pyrophoric materials. If pyrophoric materials are present in the waste, these must be processed so as to be nonflammable. Hazardous, biological, pathogenic and infectious materials in wastes must be treated so that the potential hazards from these materials are reduced as much as possible.

Requirements for gaseous radioactive wastes are also prescribed. These must be packaged so that the internal pressure does not exceed 1.5 atmospheres ($\approx$7.4 psig) at 20°C, and the total activity is limited to 100 Ci per container.

In addition to the minimum requirements on waste characteristics given earlier, minimum stability requirements are specified in 10 CFR 61.56(b). These relate to structural stability, minimization of free liquid content and void spaces in the waste. These requirements pertain to Class B and C wastes.

Structural stability means that the waste will maintain its form and physical dimensions for a minimum of 300 years under expected disposal conditions, which may include weight of overburden, moisture, microbial activity, radiation effects and chemical changes. Stability can be provided by the waste form itself or by processing to a stable form (e.g., by solidification in a binder) or by placing the waste in a container which can provide

Table 1.1.3 Concentration limits of short-lived radionuclides for Class B and Class C wastes.[a]

Radionuclide	Class B				Class C			
	Ci/m^3	Ci/ft^3	Ci/55 gal[b]	Ci/x17H[c]	Ci/m^3	Ci/ft^3	Ci/55 gal[b]	Ci/x17H drum[c]
All with $t_{1/2}$ <5y[d]	e	--	--	--	e	--	--	--
H-3	e	--	--	--	e	--	--	--
Co-60	e	--	--	--	e	--	--	--
Ni-63	70	1.98	14.5	22.7	700	19.8	145.5	227.8
Ni-63 (IAM)[f]	700	19.8	145.6	227.9	7000	198	1456	2279
Sr-90	150	4.24	31.2	48.8	7000	198	1456	2279
Cs-137	44	1.24	9.15	14.3	4600	130	957	1497

[a]Calculated from Table 2 in 10 CFR Section 61.55.
[b]55-gal = ≈7.5 ft^3.
[c]x17h drum = 11.5 ft^3.
[d]i.e., all radionuclides with half lives less than 5 years.
[e]No limits.
[f]IAM = in activated metal.

structural stability. The limits on free liquid are 1% of the volume if a container is used, and 0.5% of the volume if the waste is processed to a stable form. Void spaces in waste packages must be minimized to the greatest extent possible.

1.1.3 Effect of Regulations on Low-Level Waste Source Term Modeling - Assumptions Pertaining to Waste Inventories

It should be noted that a large part of the fundamental chemical and physical characteristics of low-level waste packages are assumed to be in accordance with NRC regulations and guidance summarized, in part in the preceeding two sections. (The NRC Technical Position on Waste Forms has not been discussed here, but it essentially recommends testing to satisfy the categorization of waste packages as "structurally stable" and includes such considerations as compressive strength, effect of moisture, microbial activity, radiation, chemical changes, etc.)

Modeling of radionuclide release rates is, naturally, heavily dependent on the particulars of the waste packages (in addition to the dependence on water flow and physical and chemical interactions that take place external to the waste packages). For the low-level waste source term modeling project, wastes will be assumed, as a minimum, to fall into the two main classes of structurally stable (Classes B and C) and not necessarily structurally stable (Class A).

In addition, several projects performed at BNL under the auspices of NRC have lead to an overview of particular wastes for a general spectrum of low-level waste generators. Specifically, fission product wastes as the result of investigations on nuclear reactor components and as the result of commercial generation of Mo-99 from fissioning of U-235 (for production of Tc-99, used in nuclear medicine) have been characterized in reports on the General Electric Vallecitios Nuclear Center and the Union Carbide Corporation [Kempf, 1984a; and Gause, 1983a]. Also, wastes resulting from commercial generation of H-3 and C-14 were investigated in a study of the New England Nuclear Corporation [Gause, 1983b], while wastes resulting from commercial production of Cs-137, Sr-90, and Po-210 sources and static eliminations were studied in a project involving the Minnesota Mining and Manufacturing Company [Kempf, 1984]. These reports represent detailed characterization of actual low-level non-stabilized (Class A) and stabilized (Class B) wastes containing a variety of radionuclides: fission products and H-3 and C-14.

Additionally, the BNL study on the Sheffield low-level waste shallow land burial site included detailed information on (and modeling of radionuclide release rates from, see Section 2.1.5 of this report) low-level wastes generated prior to the promulgation of 10 CFR Part 61. These wastes were, therefore, tremendously heterogeneous, but, based on information obtained through contacts with the original waste generators, some idea may be gained of the types of materials that might be expected to occur in Class A, B, and C waste packages (i.e., radionuclide-specific materials such as sources, research

chemical-contaminated lab trash, targets, scrubbers, decontamination wastes, etc. would still be expected to appear in low-level wastes subsequent to the issuance of 10 CFR Part 61).

In short, low-level wastes are expected to include a tremendous variety of materials. The establishment of a "representative" low-level waste trench inventory is an extremely difficult task. One simplification of low-level waste heterogeneity that may aid in the source term modeling effort, is classification into Class A (contained in carbon steel drums or wooden boxes), heterogeneous tastes (lab trash, sources, etc.) and Class B and C (contained in outer carbon steel drums or high integrity containers), stabilized and generally physically more homogeneous wastes. Generic models to accomodate outer container degradation (expected to be corrosion for carbon steel and some type of penetration/permeation for polymeric materials) and porous waste leaching should encompass a variety of Class A and Class B and C waste packages. (One such approach is presented in Section 3 of this report.) Further modeling for non-porous binder materials and non-metallic containers (as well as further modeling refinements for porous waste forms and metallic containers) will proceed as a part of the source term modeling effort.

1.2 Pre 10 CFR Part 61 Disposal Practices

Presently, there are six commercial low-level waste disposal sites: three operating and three closed. The operating sites are located at Barnwell, South Carolina; Richland, Washington; and Beatty, Nevada. The closed sites are located at Sheffield, Illinois; Maxey Flats, Kentucky; and West Valley, New York.

The design and construction of waste disposal facilities at all sites are similar. Open trenches are used as the primary burial facility with the excavated material being used as intermediate and final cover. The size of the trenches and techniques to cover the waste vary from site to site due to differences in climate and local conditions.

At both West Valley and Maxey Flats the soil underneath the excavated trenches had much lower hydraulic conductivity than within the trenches. This led to accumulation of water in the trenches and eventual water saturation within the trench. This is known as the "bathtub" effect. As the trenches filled with water, radionuclides were brought to the surface of the trenches. Rainwater incident on these trenches would come in contact with these radionuclides. Since the trench was already saturated with water, the rainwater, (now contaminated with radionuclides), would run-off along the earths' surface. Surface water run-off is a major pathway for radionuclide release for trenches that have the "bathtub" effect. At Sheffield, trench subsidence was a major problem. Before 10 CFR 61 disposal practices were such that unconsolidated, uncompacted wastes were placed in the trenches. As the waste containers degraded, the trench overburden became more than the containers could withstand. This led to subsidence and fissures in the trench caps. The fissures act as conduits for water transport to the waste and thereby can enhance radionuclide release.

The following discussion of the diposal practices prior to 10 CFR 61 at the three sites that were open both pre- and post-10 CFR 61 is abstracted from the report by Lester and co-workers [Lester, 1981] unless otherwise stated.

1.2.1 Barnwell, SC: Operating Procedures Prior to 1981

The Barnwell site has two types of trenches, "slit" trenches and "regular" trenches. The "slit" trenches handle high activity waste and the "regular" trenches are used for all other waste.

The slit trenches are approximately six meters deep and one meter wide. Their length ranges from 75 to 150 meters. During operation, a cask is lowered into the trench prior to remote removal of the inner container of waste. A crane is used to transport the container to the other end of the trench where it is immediately covered. Shipments to this trench are limited to 15,000 R/hr at the surface.

The regular trenches have a depth of 6.7 meters. Their width ranges from 15 to 30 meters. Their length varies from 150 to 300 meters. There is a 1 per cent slope from side to side. At the low side there is a trench drain system. The bottom meter of the trench is filled with sand to facilitate drainage.

The normal disposal procedure is random placement of the packaged material. Generally some effort is made to stack or position these wastes. The trenches are filled from high end to low end. In some cases, an effort is made to maintain container integrity by placing the heaviest containers at or near the bottom of the trench, although this is not always done [General Research Corporation, 1980]. Backfilling is done daily.

At the entire site, the original sand layer of approximately 1 meter thickness is replaced with a compacted clay layer. Within the trench, the waste is covered with a minimum of 0.6 meters of compacted clay followed by 1.5 to 3 meters of additional cover. When completed the area is contoured and seeded to enhance rainwater runoff.

Monitoring of radionuclide release is accomplished using wells in the trenches, around the site boundary, and off-site. Soil cores removed from initial wells extending to the water table are examined. Saturated sand layers identified in the core result in monitor wells being installed at the location of the saturated layers.

1.2.2 Richland, WA.

Trench dimensions at Richland are 7.5 meters deep, 25 meters wide and 137 meters long with some variance in length. As the climate is arid, no attempt is made to have water collection capabilities at the trench bottom. Trench construction is by dragline cranes which has the disadvantage of piling up earth on both sides of the trench leaving only the ends for a working area.

Emplacement of all waste is random and is done with a crane. There is no effort to segregate the waste. Backfilling is done as necessary to maintain doses below 100 mr/hr (typical doses are less than 5 mr/hr). High-activity wastes are occasionally received at the site. Unlike Barnwell, there are no special trenches for this waste and it is placed in the same trench as the other wastes. In fact, other wastes are often used as shielding for the high-activity waste. Backfilling is done immediately following emplacement of high-activity waste.

After the trench is filled, waste is covered with a soil cap which is approximately 1.5 meters thick at the center and 0.9 meters thick at the edges. To minimize wind erosion, a layer of cobble is placed on top of the soil.

Monitoring of radionuclide release is accomplished through daily air sampling at the site. There are no sample wells at the site, however, water, soil and vegetation sampling is performed in an extensive program on the Hanford Reservation on which the site is located.

1.2.3 Beatty, NV

Three sets of trenches were excavated at the Beatty site as of 1980. The first set, excavated between 1962 and 1965, had a trench depth of approximately 6 meters. The second set, excavated from 1965 to 1970, had a depth of 9 meters and were slightly longer than the first set. The third set, excavated from the early seventies until 1980 (the time of the site visit as reported in Lester), had a depth of 15 meters, width of 37 meters, and length of 245 meters. As at Richland, there is no attempt to collect water at the bottom of the trench since the climate is arid.

Waste is emplaced with a forklift or crane. The waste is stacked up to a height of 0.4 meters below the upper trench surface and backfilling is performed weekly. There is no segregation for high activity waste, however it is covered immediately. When the trench is full, it is covered with excavated soil with a maximum height of 1.5 meters, sloping to the sides. A gravel cover is placed on top of this soil to minimize wind erosion.

Monitoring radionuclide release at this site is accomplished with dry wells that have been drilled to a depth of three meters below the trench bottom. These wells are used for water and soil sampling. There are also two environmental air sampling stations and wells (150 meters deep) at the site.

1.3 Post 10 CFR Part 61 Disposal Practices

Major changes that occured in low-level waste disposal as a consequence of 10 CFR Part 61 revolved around the classification and subsequent packaging of the waste by the original generators, i.e., prior to shipment to the disposal site. Much of this was discussed in Section 1.1. At the disposal sites themselves the effects of promulgation of 10 CFR Part 61 appear to be dominated by requirements for generators of wastes to assure proper labeling and classification before wastes are accepted for burial. Also, segregation

of non-stabilized Class A wastes from stabilized Class B and C has been put into effect.

A brief description of the current operating procedures at Barnwell, SC is provided below. It can be seen that this is very similar to the operations prior to 10 CFR Part 61, as described in Section 1.2.1.

Trench Construction - Barnwell

Most low-level waste is unloaded into scientifically engineered trenches. All trenches are surveyed and their dimensions are documented. The floor of the trench slopes gently to the side and end where monitoring systems detect, sample, collect and remove any moisture that may enter the trench. The trenches are excavated in and capped with dense clay.

Trench sizes may vary depending on site characteristics. The trenches at the Barnwell site are generally 20 feet deep, 100 feet wide and 1000 feet long.

Waste Emplacement - Barnwell

After the truck carrying the low-level waste packages is driven to the trench, the off-loading process begins. Continuous radiation monitoring is performed through this whole operation.

Shipments arriving in vans are removed by forklift or a specially designed vacuum off-loader. Liners in shielded casks are removed by crane and placed into predesignated spaces in the trench. Waste locations are noted on a grid system in the trench and are recorded on computer files for the shipment. At the end of each day, trenches are backfilled with sand and covered with clay to prevent intrusion of moisture. Trenches that are full are mounded and capped with clay and finished off with a foot of topsoil. Grass is planted to prevent erosion, to control the runoff of rain water and to guard against seepage of water into the trench.

Permanent granite markers are placed around the trench with information on the size and contents of the trench.

Monitoring - Barnwell

Chem-Nuclear at Barnwell operates an on-site environmental testing laboratory. Health physicists and other technicians are employed to monitor and test environmental samples from the numerous monitoring points at the waste disposal site on a regular basis. In addition to radiation-measuring devices around the site, sump pipes along the edge of each trench are used to detect the presence of water and any possible migration of material within the trench itself. The sampling program monitors for contaminated materials that may leave the trench.

A cluster of three or more wells is placed at strategic locations around the site to sample water tables underground. Samples of soil and vegetation at the site, as well as radiation levels at the site perimeter, are analyzed on a routine basis.

1.4 Alternative Disposal Techniques

With only three commercial low-level waste disposal sites in operation, disposal capacity is limited and there is a need for more capacity. Due to the problems encountered at the three closed disposal facilities, a number of enhancements and alternatives to traditional shallow land burial have been proposed. Before any alternative disposal technique is found acceptable to NRC it must be demonstrated that use of the technique will result in compliance with the performance objectives in 10 CFR 61.

Five types of alternatives to shallow land burial have been identified [Bennett, 1985]: aboveground vaults; belowground vaults; earth mounded concrete bunkers (EMCB); augered holes; and mined cavities. The descriptions of these structures are abstracted from the report by Bennett et al. [Bennett, 1985] unless otherwise stated.

An aboveground vault is an engineered structure with floor, walls, roof, and limited access openings on the foundation near the ground surface. There are no constraints on the materials used to build this vault other than that the disposal system must meet the performance criteria in 10 CFR 61. Suggested materials include masonry blocks, reinforced cast in-place or sprayed concrete, pre-cast concrete, or plastics molded into solid shells. Major concerns with this concept would be the heavy reliance on the structure to meet the intruder barrier and radiation protection performance objectives.

Belowground vault disposal systems are composed of a structure built totally below the earth's surface. A belowground vault may extend above the natural surface grade provided it is covered. The advantage of a belowground vault as compared to an aboveground vault is that it will be less susceptible to climatological changes and freeze/thaw cycles. The vault could be built from a number of engineered materials such as masonry blocks, reinforced concrete, metals or plastics. The floor could be natural soil or rock, treated soil or rock, or engineered materials. The walls and roof would use engineered materials after emplacement of the waste, the vault could be backfilled to enhance structural stability.

Augered holes disposal systems are composed of shafts or boreholes that are augered or sunk by any conventional construction method that results in a cylindrical, near-surface cavity. Typical designs call for a liner that provides structural stability and resistance to water flow. The liner could be concrete, metal, or other suitable structural material. The floor of the augered hole may consist of natural soil or rock or an engineered material. Wastes would be emplaced within the shaft, covered with backfill which may be covered by an engineered material such as concrete. Augered holes have many features in common with typical shallow land burial.

Earth Mounded Concrete Bunkers (EMCB) are currently used in France. Wastes are segregated based on their level of radioactivity. Waste packages with higher levels of activity are embedded in concrete below grade and waste packages with lower levels of radioactivity are stored in metal drums and emplaced above grade in earthern mounds. After emplacement of all wastes, backfill material is placed over the entire stack. This is done to fill all voids and stabilize the earthern mound. The entire mound is covered with an impermeable clay, then with top soil and the surface is seeded. The waste disposed of above grade has an environment similar to wastes emplaced in a shallow land disposal trench.

Mined cavities include any enclosed cavity which was developed for the removal of natural resources. The Asse Salt Mine in Germany has been used for disposal of low-level radioactive wastes. In the United States, the Department of Energy and the Tennessee Valley Authority have explored the possibility of mined cavity disposal of radioactive wastes. Mined cavities present a large departure from other disposal concepts and may require special case by case modifications for licensing [Otis, 1986].

Although modeling radionuclide release rates from shallow land disposal facilities is the primary objective of this study, a secondary objective is to keep the modeling structure flexible enough to allow simple modifications for analysis of alternative disposal technologies. (The modeling structure adopted in this program is discussed in Section 3.0.) The augered hole and EMCB disposal technologies are the most similar to shallow land disposal; all three have waste surrounded by backfill covered by a cap which minimizes water infiltration. Belowground vaults are one step further removed from shallow land burial due to the engineered structure. Additional modeling work would be required to assess the performance of the structure. Aboveground vaults rely heavily on structure to meet performance requirements in 10 CFR 61. The long term stability of the structure would need to be examined with respect to climatological changes and natural phenomena such as acid rain and freeze/thaw cycles. The differences between the mined cavity concept and shallow land burial are large. Modeling release from a mined cavity may be substantially different than from any of the other proposed disposal technologies. A careful examination of the design and site characteristics would be necessary before applying the shallow land burial modeling structure to mined cavities.

1.5 Summary and Report Organization

Shallow land burial is the currently used method of disposal for low-level radioactive waste in the United States. In this technology, trenches are excavated, waste packages are placed in the trench and the trench is then backfilled with excavated soil. When the trench is full, a soil cap is placed over the trench to minimize water infiltration. This technology has been used for over twenty years.

10 CFR 61 specifies the performance objectives for any low-level waste disposal site. Before 10 CFR 61 there were no requirements on trench stability, waste form stability or on segregation of wastes based on activity. Wastes were randomly placed in the trench without concern for trench

stability. In 10 CFR 61, a classification scheme was developed which categorized the wastes as Class A, B, or C depending on the nuclide specific activity of the waste. Class A wastes are generally lower in activity than Class B or C wastes. 10 CFR 61 requires Class B and C wastes to be stabilized and segregated from non-stabilized Class A wastes. (If Class A wastes are stabilized, segregation is not required). Further, Class C wastes must be buried 5 meters beneath the trench cap to inhibit intruders from exposing this waste.

Several different technologies have been suggested as improvements to shallow land burial. These include aboveground vaults, belowground vaults, augered holes, earth mounded concrete bunkers, and mined cavities. Currently these technologies are not used in the United States, but it is likely that within the next five to ten years at least one of these techniques will be used. Currently, most interest has focused on belowground vaults, earth mounded concrete bunkers, and augered holes [Pittiglio, 1986].

Because shallow land burial is the current disposal practice, the modeling work in this project has focused on this technology. Consideration will be given to modeling release from pre- 10 CFR 61 and post- 10 CFR 61 as both types of trench exist at the operating disposal sites.

The second chapter of this report contains overviews of much of the published water flow and contaminant transport modeling work. It also contains a summary of several radionuclide release models developed at BNL for the Sheffield low-level waste disposal site. Several experimental studies are also reviewed, in particular for experiments expected to produce results applicable to the source term modeling effort: leaching tests, soil column tests, and lysimeter tests.

The third chapter discusses the compartmental modeling approach adopted in this project. This approach divides the problem of radionuclide release into four components: water flow, container degradation, waste leaching, and radionuclide transport. These components were chosen to allow flexibility in future modeling efforts which consider alternative disposal technologies since each of these processes is expected to be important for any disposal technique. Data and modeling requirements for each of these four processes are discussed. Further, chapter three provides an example of modeling release from porous wastes contained in corrodible outer containers.

A summary of the source term effort to date and discussions of projected future work are provided in the fourth chapter of this report.

2.0 MODELING AND EXPERIMENTAL WORK RELEVANT TO THE SOURCE TERM PROJECT

To provide a detailed description of the quantity and rate at which radionuclides migrate out of a disposal trench requires an understanding of the processes which lead to release. Specifically for a shallow land burial facility, release for most radionuclides will occur through transport with water. Thus, one needs to know how water moves through the trench, how waste containers degrade to allow water to contact the waste form, how waste forms leach, and how radionuclides are transported away from the waste to the disposal trench boundary. Similarly, for radionuclides that can be transported in the gas phase, one needs to know how air moves into and out of the trench, and how radionuclides enter and are transported with the air phase. Further, one needs to integrate all of these processes into a unified description of trench behavior.

There has been a substantial amount of modeling and experimental work on the various processes that lead to release. However, most of this work simplifies the problem by only considering some of the processes. For example, many models that calculate the dose to man assume the amount of radioactivity released from a trench is known a priori as a function of time. The objective of the source term modeling project is to produce a consistent model of the movement of radionuclides within a trench. This will involve use of existing models where appropriate and development of new models if none of the existing models are satisfactory.

The following sections of this chapter will provide a brief summary of some of the modeling and experimental work that is pertinent to the source term project.

2.1 Modeling

The four major processes to be modeled are water flow in unsaturated porous media, solute transport, waste form leaching and container degradation. Models are available for each of these processes ranging from sophisticated computer codes for water flow to simple empirical equations for leach rates. Also, several models that describe more than one process have been developed. Examples of these types of models are the computer codes that predict dose to man from shallow land burial. The capabilities of these models and the potential for their adaptation in this project are presented in the next few sections.

2.1.1 Water Flow Models

In predicting water flow in an unsaturated porous medium, i.e., a disposal trench, the starting point is the partial differential equation that represents a mass balance for water over the volume of the trench. For an unsaturated medium, this equation is strongly non-linear because of the dependence of material properties (such as hydraulic conductivity) on moisture content of the medium. This non-linearity and the fact that a disposal trench will not be a homogeneous medium but rather a composite of different soils and

Table 2.1 Codes identified as being useful in predicting water flow and radionuclide transport in unsaturated porous media. (Adapted from Oster, 1982 and Kincaid, 1984)

Code Name	Function	Comments
NRC-SLB	Flow/Transport	1-D*
OR-NATURE	Flow	1-D, Evapotranspiration
UNSAT1D	Flow	1-D, Evapotranspiration
SUMATRA-1	Flow/Transport	1-D
FEMWATER	Flow	2-D
FEMWASTE	Transport	2-D
TRUST	Flow	2-D
MLTRAN	Transport	2-D
UNSAT2	Flow	2-D, Evapotranspiration
VS2D	Flow	2-D, Evapotranspiration
SEGOL	Flow/Transport	3-D

* n-D, n is the number of spatial dimensions treated by the code.

waste forms, make closed-form analytical solutions difficult if not impossible to obtain. Therefore, numerical solution techniques are needed to solve the mass balance equation.

Most numerical solution techniques begin by taking the control volume and discretizing it into a number of smaller volumes that have uniform material properties. Therefore, non-homogeneous media can be easily modeled. Non-linearities can be handled through iterative solution of a linearized version of the mass balance equation.

The necessity of predicting water flow in an unsaturated porous medium has been recognized by the NRC and others that are interested in disposal of hazardous and radioactive waste. Both NRC and EPRI have had contractors conduct reviews to evaluate the computer codes available for predicting water flow and/or solute transport [Oster, 1982; Kincaid, 1984]. As a result of these reviews a number of computer codes were identified as being state-of-the-art, well documented, and available to the public. A list of these codes appears in Table 2.1. These codes differ in the number of spatial dimensions treated, the numerical solution technique, and the treatment of source/sink terms (e.g. evaporation, transpiration, etc.). As such, each code has its own strengths and weaknesses relative to the other codes.

The codes used primarily in NRC-sponsored work include: NRC-SLB [Lester, 1981], UNSAT2 [Neumann, 1974], and FEMWATER/FEMWASTE [Yeh, 1980; Yeh, 1981]. NRC-SLB uses a one-dimensional finite difference approximation to estimate water flow and was used in the system analysis of shallow land burial. UNSAT2 calculates flow using a two-dimensional finite element method; one of its strengths is in modeling evaporation and plant transpiration. FEMWATER/FEMWASTE are two codes that can be used in conjunction or independently. FEMWATER predicts water flow and provides the flow velocity used by FEMWASTE in calculating radionuclide transport. These codes use a two-dimensional finite element method and account for compressibility of the porous medium.

A more detailed description of the unsaturated water flow equation and solution techniques appears in Section 3.1 of this report.

2.1.2 Container Degradation Models

The second process leading to the release of radionuclides from the disposal trench is the degradation of the container to the point that it no longer prevents water from contacting the waste form. For Class A waste, the container may be a wooden crate which will prevent water contacting the waste for at most a few years. Class A wastes may also be packaged in carbon steel drums. For Class B and C wastes, the containers will generally have a longer life expectancy. Containers used for these wastes include carbon steel drums, high density polyethylene (HDPE), and concrete liners.

Two primary causes for a container's losing its ability to prevent water contact are mechanical and chemical in nature. Mechanical failure will occur if the container material cannot withstand the stresses imposed by the trench overburden. Chemical failure will occur as a result of interactions between the container and the soil/water/waste system.

The failure mechanism will vary depending on the material used in the container and on the local environment. For example, a steel container may fail due to pitting corrosion in an oxic environment; however, if the environment is reducing, general corrosion may be the dominant failure mode. Similarly, for a given environment, a steel may fail due to pitting corrosion while a HDPE container may suffer radiation-enhanced embrittlement which might lead to a mechanical failure. Based on literature review to determine the failure modes most likely to occur in a shallow land burial trench, models for each of the failure modes found to be important are being developed and incorporated into the source term model.

In the case of chemical degradation, the precise interactions that lead to material failure are not always well understood. For example, uniform corrosion of steels is a complex process that depends on solution Eh, pH, and major ions present, type of steel, location of defects in the steel, surface impurities, etc. For this reason, most models for corrosion are empirical in nature and take the general form:

$$X = a + kt^n \qquad (2.1.1)$$

where X is the corrosion depth, a is an empirical constant, k is a rate constant, t is time, and n is an empirical constant which gives the time dependence of corrosion depth. The values of a, k and n are determined by fitting experimental data to equation (2.1.1). Often, n is close to 1/2 which implies diffusion-controlled growth of the corrosion layer. When extrapolating corrosion depth to times much greater than the experimental times used in obtaining values of n, modelers often set n equal to 1. This provides an over-estimate of corrosion depth and is therefore conservative.

A number of models have been developed for use in predicting uniform corrosion of high-level waste containers. These models range from simple empirical expressions similar to equation (2.1.1), to more sophisticated empirical expressions in which the corrosion rate is a function of pH, and partial pressure of oxygen [Stephens, 1986] or other important variables such as oxygen and chlorine concentration [Sastre, 1986]. The most mechanistic general corrosion models predict corrosion rate through chemical reactions such as metal oxidation. This approach was used to estimate the maximum rate of uniform corrosion for steel and copper containers in an environment similar to that expected for a basalt repository [Walton, 1986].

Localized corrosion phenomena, such as pitting, are more complicated. Thus, developing a mechanistic model is more difficult than for general corrosion. Most pitting corrosion models are empirical and assume that the rate of pit growth is either proportional to the uniform corrosion rate or can be described by an equation identical in form to that used for general corrosion. That is, they use equation (2.1.1) with a larger value for the rate constant, k. The National Bureau of Standards has collected a large body of data on corrosion of irons and steels over many years which supports the use of this equation. These data have been summarized by Romanoff [Romanoff, 1957] and Campana [Campana, 1982].

Due to the complexity of pit formation and growth, there have been several attempts to predict pitting statistically. A review of the use of statistics to describe the frequency and depth of pitting corrosion of underground carbon steel can be found in the paper by McNeil [McNeil, 1986].

A model to predict the breached area of a container which has pitting as the dominant degradation mode is presented in Section 3.2.

2.1.3 Leaching

The third process leading to release of radionuclides from a disposal trench involves their release from the waste form to the contacting solution. For most radionuclides, the contacting solution will be aqueous. For some radionuclides, such as C-14 and tritium, the contacting solution may also be air. Initially in modeling the source term, consideration will be given to the species that are released to the water as this is believed to be the predominant pathway for release.

The important factors that influence release are the leachant chemistry, composition of the waste form, and system factors. A detailed literature review of how these factors influence leaching can be found in the report by Dougherty [Dougherty, 1985a]

Some of the more important factors in the leachant chemistry are the solution composition, pH, Eh, and presence of chelating agents. Leaching occurs due to a difference in the chemical potential between the waste form and the solution, thus water chemistry is important. For example, in most experiments, leach rates are higher in deionized water than in ground water. The apparent reason for this behavior is that ground waters already contain solutes which inhibit the break down of the solidification agent. The pH influences the solubility of most ions and thereby influences their chemical potential. Eh is a measure of the redox potential of the system and is an indicator of the most likely oxidation state of multivalent ions such as cobalt and the actinides. It is well known that the solubility of the actinides decreases as the Eh decreases. Chelating agents act to bind metal ions in a complex. These are often more mobile than the individual ions which tend to adsorb to the host soil. Chelating agents have been shown to lead to enhanced release of Co [Arora, 1985].

There are several materials under consideration for use as solidification agents for low-level radioactive waste. These include: cements, vinyl-ester-styrene (VES), and bitumens. Currently, most leaching studies have been done on cements as this is the most widely used low-level waste form. The choice of solidification agent has a large influence on release rates. Leach rates from cement for Cs and Sr tend to be higher than from VES or bitumens [Dougherty, 1985b]. A further difference is that cements can chemically interact with the waste whereas both VES and bitumen tend to act as inert binding agents [Dougherty, 1985b]. Even within a single class of solidification agents, the leach rate is a strong function of the waste form composition. For example, leaching of cements depends on a number of factors including: the water to cement ratio, the curing time, and the waste loading [Moore, 1977, Matsuzuru, 1979].

System factors include the temperature, pressure, radiation environment, waste form surface area to solution volume ratio, and the water flow rate and volume. Most waste forms exhibit a leach rate temperature dependence; however, because the temperature fluctuations should be small within a shallow land disposal trench, temperature effects will be ignored initially in the source term model. Similarly, pressure variations are expected to be minimal and not to influence leaching significantly and thus will be ignored. At total radiation doses expected in low-level waste, a maximum of 10^8 rads, radiation effects have a minor influence on release to the solution phase from: cements [Barletta, 1983], bitumens even though they swell and release gases [Blanco, 1966], and VES solidified wastes [Phillips, 1984]. The gases released by the irradiated bitumen may contain C-14 or tritium. However, the modeling initially will be restricted to releases in the solution phase. If radiation effects on leaching are found to be important, refinements of the models will account for this.

The flow rate or leachant renewal frequency determines the amount of time the leachant contacts the waste. At low flow rates, the concentration of some species released from the waste form may increase until their solubility limits are approached and this may limit leaching [Dayal, 1985a]. At high flow rates, there is little time for the concentrations to build up and solution feedback effects become unimportant.

Modeling waste form leaching requires identification of the mechanisms that lead to release. Values for parameters describing these mechanisms must also be assigned. Models developed have been based on some or all of the following mechanisms: diffusion, matrix dissolution, chemical interactions such as ion exchange, and corrosion. Diffusion is considered the most important mechanism and nearly every model accounts for this mechanism. All models require values for the parameters used to describe the mechanisms. These parameters are usually not obtained from first principles but from experimental data. Thus, to prevent model invalidation, caution must be exercised when selecting data from one set of experimental conditions to represent another set of conditions.

For low-level waste forms, most modeling efforts have focused on cement because it is a most widely used solidification agent. A number of attempts have been made to model Cs release from cement as a diffusion controlled process. For Sr, simple diffusion does not successfully describe release. Sr release is influenced by the presence of CO_2 and may be incorporated into the cement matrix. The release of actinides and Co are often found to be solubility-limited. Very few mechanistic models have been developed to describe the release from VES or bitumen.

Due to the interest in glass as a high-level waste form there has been a great deal of modeling work on borosilicate glass leaching [Pescatore, 1983; Mendel, 1984]. However, much of this work is not directly applicable to low-level waste forms. For example, glass is an impermeable solid whereas concrete is a porous solid. It is possible that some of the concepts applied to modeling glass leaching can be modified and applied to low-level waste form leaching.

In summary, leaching is a complicated process. Development of any model will require consideration of the properties of the solidification agent, of the radionuclide being modeled, as well as of the leaching solution.

A model of release from a porous waste form is presented in Section 3.2.

2.1.4 Radionuclide Transport

The fourth process leading to release of radionuclides from a disposal trench involves their transport from the waste form to the trench boundary. With the exception of radionuclides such as C-14 and tritium which can form gaseous species, transport will occur through the water. Important factors that influence radionuclide transport include: diffusion, dispersion, advection, and chemical interactions with the soil.

Diffusion is a result of the random molecular motion of the radionuclides. If the concentration of radionuclides were constant throughout the system, the random motion of the molecules would not lead to a change in concentration. However, if there were a region of high relative concentration, the random motion would cause a gradual shift in concentration from the higher region to the lower region until there were no concentration gradients in the system. Diffusion will occur in a moving or stationary fluid.

Dispersion is caused by the variations in fluid velocity within the pore spaces. Velocity variations are caused by variations in pore size and geometry, the velocity profile within the pore, and tortuosity. For the radionuclides being transported with the water, the velocity variations lead to some radionuclides being transported faster than others.

Advection refers to the movement of the radionuclides along with the flowing groundwater. The rate of advection is equal to the pore water velocity which is defined as the velocity given by Darcy's law divided by the kinematic porosity, the volume of flowing water per unit bulk volume of porous medium. The advection velocity is the mean velocity with which the water travels. In practice, there are variations in fluid velocity within the porous medium as discussed above. These variations are modeled through the dispersion concept.

Chemical interactions between the soil and water include: adsorption-desorption, ion exchange, and precipitation-dissolution. Other chemical reactions that will influence transport are colloid and complex formation as well as microbial interactions. Increased transport rates of Cs, Sr, and Co have been attributed to chelation by EDTA [Arora, 1985]. Chemical reactions define the distribution of radionuclides between the solid and solution phases and are a strong function of the water chemistry (e.g., pH, Eh, etc.).

Modeling of radionuclide transport away from the trench to the accessible environment has received substantial attention in predicting dose to man from simulated shallow land burial sites [Lester, 1981; Hung, 1983; King, 1986]. For processes occurring within the trench there has been little modeling work. However, attempts to understand and predict migration in column studies [Ohnuki, 1986] and lysimeters have been made [Polzer, 1985; Wilhite, 1986].

The starting point for modeling contaminant transport is the advective-dispersion equation which treats the four processes previously discussed. In this approach, chemical interactions are treated as a source/sink term. The most rigorous method of handling the chemistry is to include models for all of the chemical interactions for each species. This requires an extensive geochemical data base and expands the number of equations that must be solved in the system since each radionuclide compound requires its own transport equation. A widely used approximation for the chemical interactions term is to assume that each radionuclide is reversibly sorbed to the soil in direct proportion to its solution concentration.

A more detailed discussion of modeling radionuclide transport within the disposal trench is presented in Section 3.3.

2.1.5 Sheffield Waste Radionuclide Release Modeling Work

The Sheffield, Illinois, low-level waste burial site, which was closed in 1978 has been considered by NRC to be capable of yielding data of value for NRC's regulatory purposes of serving, as it were, as a laboratory for obtaining information applicable to other disposal sites, as well as for developing a predictive model of the Sheffield site behavior. Both these functions required detailed knowledge, not only of the amounts and isotopic composition of the buried waste, but also of the waste forms and packaging. These last are essential for estimating release rates of the various isotopes from the waste, which was a specific aim of the Sheffield study.

At the Sheffield burial site, tritium had been found in several places outside the trenches since shortly after the site was closed. In at least one area, it had also been found in small amounts (well below the maximum permissible concentration) at a considerable distance outside the site boundary. It was thus important to try to estimate the amount of H-3 which was likely to be released in the future, and the rate of its release. The same was true for all isotopes of reasonably long half-life ($\geq$5 yrs). Inventory estimates for this site were in wide disagreement prior to the initiation of the BNL study. Thus, one of the main goals was to prepare an accurate inventory of H-3 and C-14 (as well as other selected isotopes of half-life >5 yrs) for eight of the Sheffield trenches. The waste inventories are, of course, a minimum requirement for estimating isotopic release rates from the trenches.

An inventory was made of the contents of Trenches 1,2,7,11,14A,23,14, and 25C at the Sheffield LLW burial site. For this purpose, microfilm copies of the radicactive shipment records (RSRs) were reviewed. Using the RSRs, compilation was made of the amounts of relevant isotopes with half-life >5 yrs shipped to each trench of concern. The compilation was done with the help of a data base set up on BNL's CDC 6600 computers using Intel Corporation's System 2000 data base management system. Information from some 1700 non-fuel cycle RSRs and 3200 fuel cycle RSRs was stored [MacKenzie, 1985].

On the basis of information supplied by nuclear power plant operators on isotopic composition of their waste, estimates of the trench inventories of fuel cycle Cs-137 and Co-60 were made. The Sheffield site was operated prior to the establishment of the LLW A,B, and C classification system and performance objectives set up in 10 CFR Part 61; thus, the waste packages at the Sheffield site represented a very heterogeneous mix of low-level wastes. Solidification of liquid wastes was a general practice, but the concept of a monolithic waste form or a stabilized waste had not been thoroughly developed nor systematically applied.

It was found that there existed a wide divergence in the non-fuel cycle shipments to the various trenches, both in isotopic distribution and in waste category. This was due to several factors, such as changing research programs at institutions and changing business ventures of industrial firms. Fuel cycle waste is generally much more uniform in terms of isotopic composition

and waste type, and variation in amounts among trenches was not generally great because of the large volumes shipped to all trenches.

In connection with the modeling done to estimate release of activity to the trenches, exact descriptions of waste forms and containers were often not available, and assumptions had to be made. These assumptions were made conservatively so as not to underestimate possible releases. At the same time, an attempt was made to keep the assumptions realistic. Estimates of isotopic release rates involved modeling the behavior of a number of different container-waste form combinations under the conditions found in the trench environment. Both containers and waste forms of standard types, and those which were non-routine, were considered.

Information on waste form was rarely given on non-fuel cycle RSRs. This was obtained for shipments of relatively large amounts of activity by contacting the generators. These generators accounted for roughly 90% of the H-3 waste in most trenches. In certain cases, information on special containers was also obtained.

Fuel cycle waste information which was not always supplied on the RSRs and which was required for modeling, included whether or not the waste was solidified, and the nature of the solidification agent. A detailed study was made of one trench (Trench 24) to obtain a breakdown into the amounts of the different isotopes (Co-60, Cs-137, and Sr-90), which were contained in unsolidified waste and in concrete and urea formaldehyde resin, in either drums or liners. The proportions arrived at were applied to the other trenches. Summaries of the modeling done for H-3, C-14, Sr-90, Cs-137, and Co-60 are presented in the following sections.

2.1.5.1 Tritium Waste Release Modeling

Low-level wastes containing tritium that were buried in the eight trenches studied at Sheffield included a wide variety of waste forms and represented an approximate total inventory of ≈2340 Ci. Of this total, ≈40 Ci were fuel cycle waste while the vast majority of the wastes (containing ≈2300 Ci) were non-fuel cycle. The types and actual physical and chemical form of the tritium waste had to be determined through contact with the original generators since the RSR investigation yielded extremely little information on specific waste characteristics. There were essentially eight broad categories of tritium waste:

- Packages in shipments < 1 Ci solid
- Packages <1 Ci liquid, packages >1 Ci liquid, and solid packages >1 Ci general laboratory trash shipped in drums
- Targets (tritium or zirconium tritides)
- Scrubbers (T_2O sorbed on desiccant)

- Tritiated organics, particularly luminous paint
- Tritiated cement
- General laboratory trash
- Packages >1 Ci, all activity released during first year (e.g., HTO in vials, broken glass bulbs, and packages >1 Ci shipped in fiberboard boxes).

In modeling the release of tritium from these various types of waste, the modelers had to take into consideration several mechanisms or processes having the potential to contribute to the degradation of the waste package and/or to lead to "direct" release. These processes included:

- diffusion - considered for movement of water vapor through concrete,
- permeation - considered for liquid water movement through concrete as well as for tritiated water and tritium gas (HT or T_2) through polyvinyl chloride and other polymers and through glass,
- radiolysis - considered for production of HT/T_2 gas from HTO/T_2O,
- exchange - considered for release, particularly in the gas phase, based on exchange of tritium for hydrogen in water vapor.

Release rates of tritium from targets, luminous paints, and from concrete blocks were modeled based on experimentally determined data. The release from target materials was found to follow a curve of relatively high initial percentage release followed by a smaller steady percentage release thereafter. The tritium release from luminous paints had been found to follow first order kinetics (first order in tritium). Contaminated lab trash was considered to release its tritium in a manner analogous to loss from luminous paint and thus, first order kinetics were applied to the packages containing lab trash.

Concrete blocks containing tritium (tritiated cement) released tritium at a rate which could be fitted to an expression with time dependence $t^{1/2}$. A diffusion mechanism of radionuclide release was inferred from this behavior.

Throughout the modeling (independent of the waste form), the process of radiolysis production of HT/T_2 gas had to be considered as well as the potential exchange of tritium (no matter what its chemical state) with hydrogen in ambient water vapor.

Distinguishing characteristics of tritium waste that make modeling its release rather complex include its ability to exchange with hydrogen.

Hydrogen may be present in low-level waste in many forms; the most abundant is expected to be water (either as liquid or as vapor). Very pertinent to radionuclide release considerations is the fact that water may be physically present as water of hydration (in concrete or as part of hydrated salts/solids), as ambient water vapor, or capillary or adsorbed water throughout the soil, or as "bulk" water traveling through the waste in a wetting front. There is a potential for tritium to exchange with the hydrogen of any of these forms. There is, additionally, the possibility that exchange for hydrogen in organic or polymer molecules could occur. Tritium has also been reported as not readily sorbed on or dispersed through soils [de Sousa, 1985].

2.1.5.2 Carbon-14

The low-level waste C-14 inventory in the eight trenches studied at Sheffield totaled ≈104 Ci. Just over half of this amount was solid organic C-14-labeled material and the remainder was miscellaneous C-14-contaminated waste including paper, glassware, animal carcasses, gloves, etc. The solid organic C-14 labeled wastes were modeled to release their radionuclide activity by two general mechanisms:

- gaseous releases from C-14 radiolysis, and,
- release by contact with water through
  - simple "loss-on-contact" leaching,
    and
  - waste volume removal "washing."

The C-14-contaminated lab trash was modeled to release its activity by the simple contact-loss leaching mechanism. It was felt that radiolysis production of gases in such materials would be minimal due to the low specific activity of C-14 and concurrent ineffective dispersal of emitted radiation (ineffective from the point of view of radiolytic gas production).

For the solid organic materials, the gaseous releases from C-14 radiolysis were coupled to the simple contact-loss leaching in such a way that the source term available for radiolysis and leaching was diminished simultaneously by these two mechanisms. The radiolysis calculations were based on conservative G-values and the assumption of total radiation absorption. The contact-loss leaching represented an approach in which the C-14 present in a particular region of a drum leached as soon as the drum corroded in that area. The weighting factor applied to the C-14 source term was the ratio of corroded area to non-corroded area. In other words, a 30% corroded drum would have cumulatively released 30% of its C-14 source term by leaching. As mentioned, this was the approach used for modeling releases from the miscellaneous C-14-contaminated wastes as well.

The waste volume removal "washing" model represented a case in which incoming water volumes (limited by corrosion area in the outer drum) actually

"washed-out" an equivalent volume of waste. The wastes were assumed to contain homogeneously distributed radioactivity and the released material was uninhibited in its exit.

2.1.5.3 Sr-90, Cs-137 and Co-60

Non-Fuel Cycle Sr-90, Cs-137 and Co-60

Non-fuel cycle inventories for Cs-137 and Co-60 were small compared to fuel-cycle contributions except in two trenches. These contained non-fuel cycle Sr-90 and Cs-137 as soluble salts and on labeled microspheres packaged in lead-lined drums and generated by the 3M company. The totals were ≈2000 Ci of Cs-137 and ≈1300 Ci of Sr-90. In fact, the non-fuel cycle Sr-90 amounts in these trenches were comparable to the fuel cycle Sr-90 inventories in other trenches.

The model developed for the 3M Sr-90 and Cs-137 wastes took into consideration several material and process factors, including:

- the carbon steel outer container,
- the lead lining inside the carbon steel,
- the possibility of a galvanic couple between the steel and the lead,
- the dependence of radionuclide release amounts on orientation or position of the waste package in the trench,
- the possibility of radiolysis production of gases, and of acids which might contribute to corrosion of the lead lining,
- the variation in rain water amounts which might be incident on the container, and
- the feasibility of diffusion-limited modeling for the radionuclide releases.

The final model developed for these wastes represented a three-phase sequence. The first phase involved an induction to first pitting of the outer container and then restricted influx of water through the pit until an opening the size of the drum top had corroded away. This allowed water influx through the crack around the lead-lining lid. The second phase consisted of build-up of water in the lead lining and, concurrently, dissolution of the Sr-90 and Cs-137 materials. At the end of the second phase, the waste package consisted of a corroded outer container and an inner liner containing a "bathtub" of dissolved material with an upper fluid level even with an exit crack or indentation for the lead lining lid. The third phase consisted of repeated mixing and removal of incoming rainwater "rinses" with the "bathtub" solution

standing in the lead lining. By this process of successive removal of solution, the radionuclide releases from these wastes were calculated.

Fuel-Cycle Sr-90, Cs-137 and Co-60

Fuel-cycle inventories of Cs-137 and Co-60 were estimated at ≈5900 and ≈6300 Ci, respectively. The fuel-cycle Sr-90 inventory was taken as ≈10% of the Cs-137. These radionuclides occurred in two main types of waste:

- cement- or urea-formaldehyde-solidified wastes in carbon steel drums or liners, and
- unsolidified waste in carbon steel drums or liners.

The basic approach applied to these wastes was one in which the infinite plane sheet solution data given in ANS-16.1 (Working Group ANS 16.1, 1982) was digitized and fit with a quadratic power series with

$$CFR(t) = C_0 + C_1X + C_2X^2$$

where $X(t) = \frac{S}{V}\sqrt{Dt}$.

In these expresions, C_o was set equal to zero,

C_1 = 1.3441,

C_2 = - 0.4416,

S = surface area of the waste form

V = volume of leachant

D = diffusivity of radionuclide.

The diffusivities used for the solidified waste radionuclides were assumed to reflect the order of magnitude values for these isotopes consistent with leaching data in the literature [Colombo, 1979; Dayal, 1983; Haggblom, 1979].

Also, the corrosion area of the outer drum was included as an effective surface area ratio, i.e., corroded area divided by total area. This factor was multiplied with the CFR calculated earlier and resulted in limiting the available "source" for leaching.

Unsolidified trash waste was modeled with a similar approach except the release function was modified to make the release at any time directly

proportional to the exposed surface area at that time and to the fraction of material remaining in the container at that time.

2.1.6 System Models

The sections (2.1.1) through (2.1.4) discussed the modeling of the four processes that lead to radionuclide release from a low-level waste disposal trench. Section 2.1.5 discussed particular radionuclide release models from waste packages. These models tended to focus on a single aspect of release and not on the entire system. There has been some simplified system modeling of low-level waste disposal sites. NRC contractors developed a code used in the assessment of shallow land burial systems [Lester, 1981] and more recently have developed the code Onsite/Maxi-1 [Kennedy, 1986]. EPA and their contractors have developed the PRESTO-EPA computer code [Hung, 1983]. DOE contractors at Savannah River Laboratory developed the DOSTOMAN code [King, 1986]. However, these models treat release from the trench in a simplistic manner. None of these analyses model container degradation. Also, leaching is defined through input as an annual fractional release rate or, in the DOE model, as an exponentially decaying release rate.

Systems models that consider container degradation and waste form leaching have been developed for analyzing proposed high-level waste repositories in the United States and Canada as well as intermediate-level waste repositories in the United Kingdom. Respectively, these models are WAPPA [Intera, 1983], SYVAC [Dormuth, 1981], and VERMIN [Electrowatt, 1983]. These models are written for saturated environments and as such are not applicable to shallow land burial. However, the metallic corrosion models and leaching models in these codes are general enough that they may be useful, provided data relevant to shallow land burial is used in evaluating the rate parameters.

2.2 Experimental Work

Experiments geared towards developing an understanding of the processes that may lead to radionuclide release from a disposal trench tend to focus on waste form leaching and radionuclide transport in unsaturated porous soils. Water flow is studied in relation to transport of the radionuclides.

The three major experiment categories are: leaching experiments which provide information on the interaction of the waste form and solution; column tests in which a tracer is injected at the top of the column, in these tests both water flow and transport in porous media are examined; and lysimeter tests which involve a waste form surrounded with a porous soil, these tests obtain information on water flow, leaching, and transport.

The following sections provide a brief description of these types of tests and discuss how their results may be applied in development of the source term model.

2.2.1 Leaching Experiments

In a typical leaching experiment, a waste form is loaded with trace amounts of radioactive material and then submerged in an aqueous solution. After a period of time, the leachant is analyzed to determine the amount and identity of the leached material. Often, a surface analysis of the waste form is performed to complement the solution analysis.

There are two basic categories of leach test: static and dynamic. These tests evaluate leaching behaviors for stagnant and flowing leachant, respectively. Tests which periodically replace the leachant are considered dynamic. Many different test procedures have been used or proposed [MCC, 1981]. Two widely used dynamic tests for low-level waste forms are the procedures recommended by ANS [ANS, 1984] and IAEA [Hespe, 1971]. Both tests require that the leachant be replaced periodically. The main difference between these tests is in the choice of times at which the leachant replacement occurs.

Leaching tests have been performed on the three types of solidification agent for a wide variety of radionuclides. The majority of the work has been focused on leaching of different types of cements. The most frequently studied radionuclides are Cs, Sr, and Co. However, leaching of other radionuclides including tritium, C-14, Pu, Am, I, and Mn has also been studied.

The general experimental results of leaching of cement is that all species have a rapid initial release. This is attributed to a surface wash-off effect. After an initial period, releases occur at a slower rate. For Cs, Sr, and Co; Cs release occurs at a faster rate than Sr which is released at a faster rate than Co. The most common explanation for this behavior is that Cs exists in the pore waters of the concrete and its exchange is limited by the diffusion in solution. Sr is believed to become incorporated into the concrete matrix structure and thus, its release is related to the dissolution of the matrix. Co release is believed to be limited by its low solubility in the high pH environment characteristic of concrete waste forms.

In leaching experiments in which VES was used as the solidification agent, cumulative fractional releases (CFR) of Cs, Sr, and Co were nearly identical. This indicates that none of these species interacted with the VES [Dougherty, 1985b]. The effects of temperature on leaching from VES waste forms was also studied. The release rates of Cs, Sr, and Co were measured at temperatures between 20°C and 70°C. There was no consistent pattern in the change of leach rates with temperature: the release rate of 30 °C exceeded that at 50°C but was less than the release rate at 70°C. Again, there was little variation between Cs, Sr, and Co in terms of release [Dougherty, 1985b].

Leaching experiments on bitumen yielded CFR's of Cs, Sr, and Co which were identical within the limits of the reproducibility of the experiment. The leach rate of bitumen did not change as the temperature increased from 20°C to 50°C. The reason for this is believed to be that radionuclides are held in the bitumen as coated salt particles. Thus, the permeability of the

bitumen is the controlling factor in release and it does not appear to change greatly over the temperature range tested [Dougherty, 1985b].

Another category of leaching tests involves wet/dry cyclic leaching. The static and dynamic leach tests described occurred under water-saturated conditions. However, in a low-level waste disposal trench, the soil will be unsaturated with respect to water and there may be long periods in which precipitation is low and the soil will tend to lower degrees of saturation. In order to simulate this type of environment more closely, wet/dry cyclic leaching experiments were performed [Arora, 1986]. In these experiments, cement waste forms were allowed to leach in a static solution for a specified period. After this time, the waste forms are removed from the water and held in a dry environment for a few days. This cycle was repeated for the duration of the experiment. Results of these experiments indicate that the total release from the waste forms as a function of time was always less for the cyclic leach tests as compared to the saturated leach tests. However, after a dry period, the release rate of Cs increased to a value greater than at the end of the previous wet period. This was attributed to Cs diffusing to the pore surfaces of the concrete and becoming readily available for release as soon as the next pulse of water contacted the waste form. In contrast, Sr release after a dry period occurred at a slower rate than at the end of the previous wet period. The reason for this is believed to be incorporation of Sr into the concrete as it cures. Due to the absence of soil, which would retain some moisture even in the absence of precipitation, wet/dry cyclic leaching experiments do not accurately reproduce the expected field conditions. Similarly, saturated leach tests do not reproduce the expected field conditions. Due to the differences between wet/dry cyclic leaching results and those from saturated leaching experiments, it will be necessary to determine the conditions under which each of these types of experiments is relevant to modeling release from a shallow land burial facility.

2.2.2 Column Tests

Column tests have been performed to determine how soils will interact with radionuclides released from low-level waste forms. In a typical column test, a column is packed with soil, and water (spiked with a tracer) is injected at the top and allowed to flow through. By monitoring the effluent at the bottom of the column, a measure of the interactions between the soil and the tracer can be obtained.

In general, anions and neutral species tend to be non-reactive or weakly reactive with the soil. In contrast, cations tend to interact with the soil and become adsorbed to the soil surface, thereby slowing their transport. For example, a large fraction of Cs in solution will sorb to most soils. Thus, even though its release rate from the waste form may be large, its transport to the accessible environment will be slow provided it does not form a chelated compound.

Column tests have been done for a number of species; examples of the types of behavior that might occur in a shallow land disposal trench might be inferred from qualitative results such as those that follow. Cs is strongly

sorbed by most soils and will not migrate at a rapid rate [Dayal, 1985b]. Sr is sorbed to some degree by soils and its transport is much slower than water but faster than Cs [Stone, 1986]. Co has been found to join complexes which do not interact with the soil and thus the transport of Co can be more rapid than Cs or Sr [Oblath, 1985]. Am can become part of colloidal particles which interact with the soil less frequently than elemental Am. Thus, the primary method of Am transport through glauconitic sand was found to be with colloidal particles [Saltelli, 1984].

The degree to which colloids form or radionuclides sorb to the soil is highly dependent on the local environment (i.e., water chemistry, soil type, presence of chelating agents, etc.). Thus before using quantitative data to support modeling of radionuclide transport, it must be insured that the data is relevant to the situation being modeled.

2.2.3 Lysimeter Tests

Lysimeter tests are performed to reproduce the actual conditions expected in a shallow land burial facility as closely as possible. To achieve this, a water-tight cylinder open at the top and bottom is surrounded by and filled with soil. The source of water is usually rainfall minus evaporation and transpiration (if any) at the top surface. However, in some instances water is introduced at the surface at a specified rate. Drainage is allowed to occur through the bottom surface of the lysimeter. To simulate leaching, a waste form is placed beneath the soil surface. In some cases, vegetation is planted at the soil surface in order to allow study of radionuclide uptake by plants. In other cases, the top of the lysimeter is filled with different soil layers to simulate a trench cap.

Lysimeter studies provide information on water flow and the spatial and temporal distribution of contaminants beneath the waste form. This data is obtained through analysis of water samples and soil cores. Water samples are obtained from porous cup samplers located at various distances from the waste form or from the effluent at the bottom of the lysimeter. At the end of the experiment, soil cores are often taken to provide information on spatial distribution of contaminants on the soil.

Two major differences between the environment in the lysimeter studies and in the expected disposal conditions are the absence of a container and the absence of interactive effects that may occur as a result the water chemistry modifications which occur through contact with other wastes and waste forms.

Table 2.2 is a partial listing of lysimeter studies being performed in the United States. This listing represents the majority of the work reported in the last five years. In the cases where several reports have been prepared over the years for a particular lysimeter only the most recent reference is cited. From the table, it can be seen that in some tests actual low-level wastes from power reactors are being used. This makes the tests more representative and insures that a wide range of contaminants are studied.

Table 2.2 Partial compilation of lysimeter studies.

Location	Primary Contaminants	Waste Form	Comments	Reference
Los Alamos	Li, I, Br, Cs, Sr	None	Spiked incoming solution	Polzer, 1986
Savannah River	Mn, Co, Sr, Ru, Sb, Cs	Lab trash	7 year tests*	Stone, 1986
Savannah River	Pu-238, Pu-239, Am-241	Defense Waste	2 year tests*	Stone, 1986
Savannah River	Tritium	Stainless Steel Crucibles	12 year tests	Stone, 1986
Savannah River	Tc-99, Nitrate	Saltstone	Defense Wastes- 3 year tests*	Wilhite, 1986
Savannah River	Mn, Co, Zn, Sr, Cs, Ce	Portland Cement Masonry Cement VES	Power Reactor Wastes 3 year tests*	Oblath, 1985
PNL	Mn, Co, Sr, Cs	Portland Cement Bitumen VES	Power Reactor Wastes, 1 year test*	Skaggs, 1986
PNL	None	None	Moisture Migration	Jones, 1978
Maxey Flats	None	None	Moisture Migration	Schulz, 1986
Georgia Tech	None	None	Moisture Migration	Eicholz, 1985

* Length of tests are as reported in the reference, these tests are continuing.

Some interesting results of the lysimeter tests include the observations made at Savannah River regarding the leaching of nitrates from saltstone. In comparing leaching of nitrate from saltstone submerged in water and saltstone surrounded by unsaturated soil with approximately 20% (by volume) water, they found the leach rates were nearly identical. Further, no effect of moisture content on leach rate was observed until the moisture content was reduced to 1% [Wilhite, 1986]. A moisture content of 1% is far below the expected value in a humid climate. The second interesting result from these tests is obtained from comparison of the results between uncapped and capped lysimeters. In the uncapped lysimeter, measurable amounts of nitrate and Tc have been found in the effluent at the bottom of the lysimeter. This is not the situation in the capped lysimeters. In the capped lysimeters there was very little water released at the bottom. However, soil moisture samples from regions adjacent to the waste form in the capped lysimeters show significant concentrations of nitrate and Tc. This supports the results discussed earlier in this paragraph that leaching may occur even in the absence of significant water flow.

These results indicate that, in this case, for soluble species, results of standard laboratory leaching tests may be adequate to describe release from a waste form buried in an unsaturated porous medium. Also, although trench caps can be effective in reducing water flow to the waste form, this did not stop leaching of soluble species in this experiment. If at some later time, the trench cap loses its ability to reduce water flow, these soluble species will be in a position to be readily picked up by the flowing water and this may lead to a large spike in the release. This will be a particular concern for long-lived species such as Tc-99.

In tests in which the lysimeter surface is covered with vegetation, uptake by plants has been shown to be a significant pathway for movement of Cs and Sr [McIntyre, 1986] and tritium [Schulz, 1983].

The lysimeter tests should most closely represent the expected conditions within a shallow land burial facility. As such, it is hoped that they will provide useful data for source term modeling and model validation.

2.2.4 Container Degradation

The performance of the container has not received much direct attention in relation to its use in low-level waste disposal. However, for steels and concretes, there is a large data base of information on their properties and performance in other fields. The summary of data on underground corrosion of steels by Romanoff [Romanoff, 1957] and Campana [Campana, 1982] should be useful in model development. An assessment of the performance of concrete as a structural material for alternative low-level waste disposal technologies has been performed [MacKenzie, 1986]. Many of the properties that would influence the ability of a concrete container to retard radionuclide release would be similar to the properties that make concrete useful as a structural material [e.g., resistance to water flow].

High density polyethylene (HDPE) is another material being suggested for use in shallow land burial. The data base on HDPE properties is not as great as that for steels or concretes and there is concern about guarantees of maintenance of integrity of an HDPE container for 300 years under actual burial conditions. In an attempt to address some of the concerns about the performance of HDPE, studies have been conducted to investigate the effects of environment and gamma irradiation on its mechanical properties [Soo, 1986].

2.3 Conclusions

Modeling of radionuclide release from low-level wastes in a shallow land burial facility will be achieved through considering four processes: water flow in unsaturated porous media, container degradation, waste form leaching, and radionuclide transport. A considerable amount of modeling work has been done on water flow and contaminant transport. There are several computer codes that calculate these processes and some of these codes should be adaptable to the needs of the source term program. Much less work has been published in the literature on modeling of container degradation and leaching; this is particularly true for bitumen and VES waste forms, and HDPE

containers. If the available models are found to be non-existent or inadequate, new models will be developed in the source term project for these processes.

The primary emphasis of experimental work in low-level waste management has been on leaching and radionuclide transport in unsaturated soils. In the laboratory, leaching of waste forms has been studied using methods similar to the IAEA and ANS 16.1 test standards. Radionuclide transport has been studied through packed soil columns. In the field, lysimeter studies are widely used to measure moisture migration, waste form leaching, and radionuclide transport under conditions similar to those expected in a shallow land burial facility. The data from these experiments should be useful in obtaining the parameters used in the models for leaching and transport. The results of these experiments (particularly the lysimeter tests) should also be useful in validating the source term models.

3.0 MODEL DEVELOPMENT

NRC is interested in developing the capability to predict the rate of radionuclide release from low-level waste disposal sites. This includes shallow land burial as well as alternative disposal techniques that are currently being proposed. In the United States, up to the present time all low-level waste disposal sites have used shallow land burial. For this reason, initial modeling efforts will be structured towards calculating radionuclide release from a shallow land disposal trench. For a system as complicated as a shallow land burial trench, realistic simulation will require computer modeling.

A primary objective of the modeling effort is to make the solution procedure flexible enough to allow incorporation of new models to represent alternative disposal methods while retaining the basic procedures used for modeling shallow land burial. This will be accomplished by structuring the computer code to consist of a series of modules that represent the major physical processes that influence disposal site performance. These modules will be composed of a series of models that represent the mechanisms that influence the physical process under consideration.

The first step in developing a model is to define the system to be modeled. This can be accomplished through identification of: the major physical processes involved in disposal site performance, the materials used in disposing of the waste, and the mechanisms that influence performance.

In this report, a major physical process is defined as a series of gradual changes, possibly caused by several different phenomena, that influence site performance. The major physical processes have been identified [Sullivan, 1985] as shown in Figure 3.0.1: water infiltration, container degradation, waste form leaching, and radionuclide transport from the waste form to the edge of the disposal unit. These four processes will form the basis for modeling low-level waste disposal sites.

In the next stages of model development, specialization to a particular system is required. The system selected is shallow land burial.

The materials present in shallow land burial can be divided into four categories; structural materials, containers, waste forms, and waste. The structural materials include all of the components used in forming the trench such as trench caps, liners, and backfill. The containers include cements, high density polyethylene plastics, and carbon steel. The waste forms include cements and bitumens. The waste refers to the radionuclides that are incorporated into the waste form.

The mechanisms that may affect radionuclide release are identified for each material through review of the literature. For example, pitting corrosion of carbon steel drums is one mechanism that can lead to the container's losing its ability to prevent water contact with the waste form. After the mechanisms have been identified, the physical parameters and data

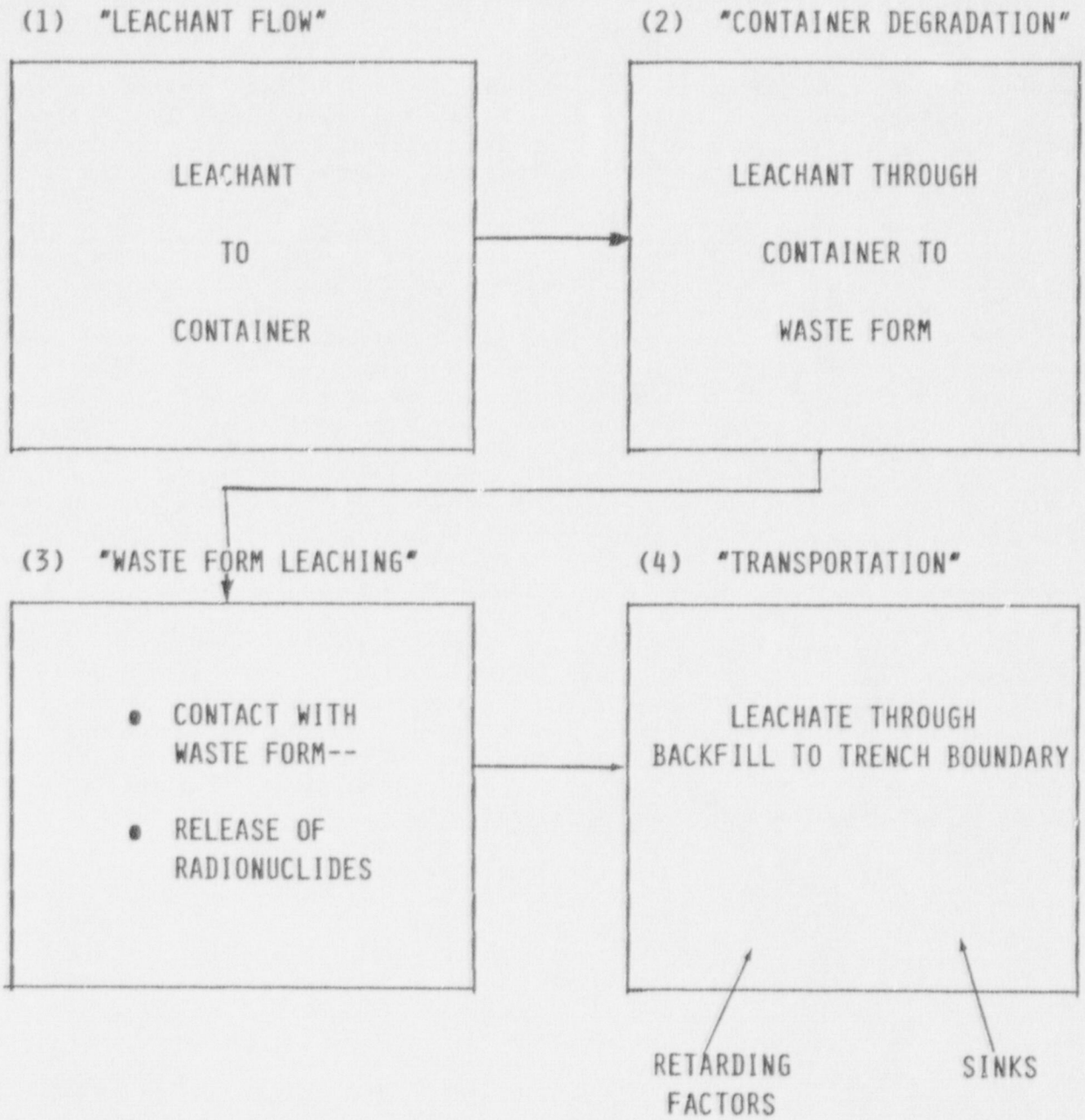


Figure 3.0.1 Process modules for representing a low-level waste disposal unit.

necessary to quantify the rate with which the mechanism proceeds can be identified. For pitting corrosion, the rate of pitting is the important parameter and this is influenced by the particular material, temperature, water chemistry, and other environmental parameters.

Figure 3.0.2 illustrates the concepts presented in the preceding paragraphs. The four process modules are expressed across the top of the figure. In this figure, for example, attention is focused on container degradation where the material identified is carbon steel. The mechanism affecting performance is pitting and the rate of pitting is the important parameter. This pitting rate will be a function of the local environment.

Figure 3.0.3 is an extension of the ideas presented in Figure 3.0.2. In this figure, the module representing container degradation is expanded in more detail. The top line lists the materials considered for containers. The next line lists the mechanisms that influence container performance. Each of these mechanisms will require a separate model. The next line lists the rate parameter which will be the result of the models of the mechanism directly preceding in the chart. All of the models will be influenced by the local environment as expressed in the last line of the diagram. The diagram is not meant to be a complete listing of all container materials or degradation mechanisms, however it is meant to give a flavor for the solution procedure and the complexities involved due to the variety of materials used in low-level waste disposal. Similar diagrams could be presented for each of the three remaining process modules.

Figure 3.0.4 is a flow chart for the solution procedure that will be used in solving for the rate of radionuclide release from a low-level waste disposal site. The left side of the flow chart contains symbolic representations for the events that define the problem and control the flow of the calculation. In particular, the problem is defined through user supplied input and initialization. After completion of these steps the calculational time will be started and the initial state of the system will be given as output. From this point, the main part of the computation will be conducted. This is represented by the four process modules on the right side of the flow chart. The calculation will proceed sequentially by calculating the water infiltration, container degradation (if all of the containers are intact the next two steps will be skipped and the time will be updated), waste form leaching, and radionuclide transport for a given time step. At this point, the calculational time will be increased and the process repeated.

In the next few sections, the development of background information and of considerations pertinent to modeling the four process modules is presented.

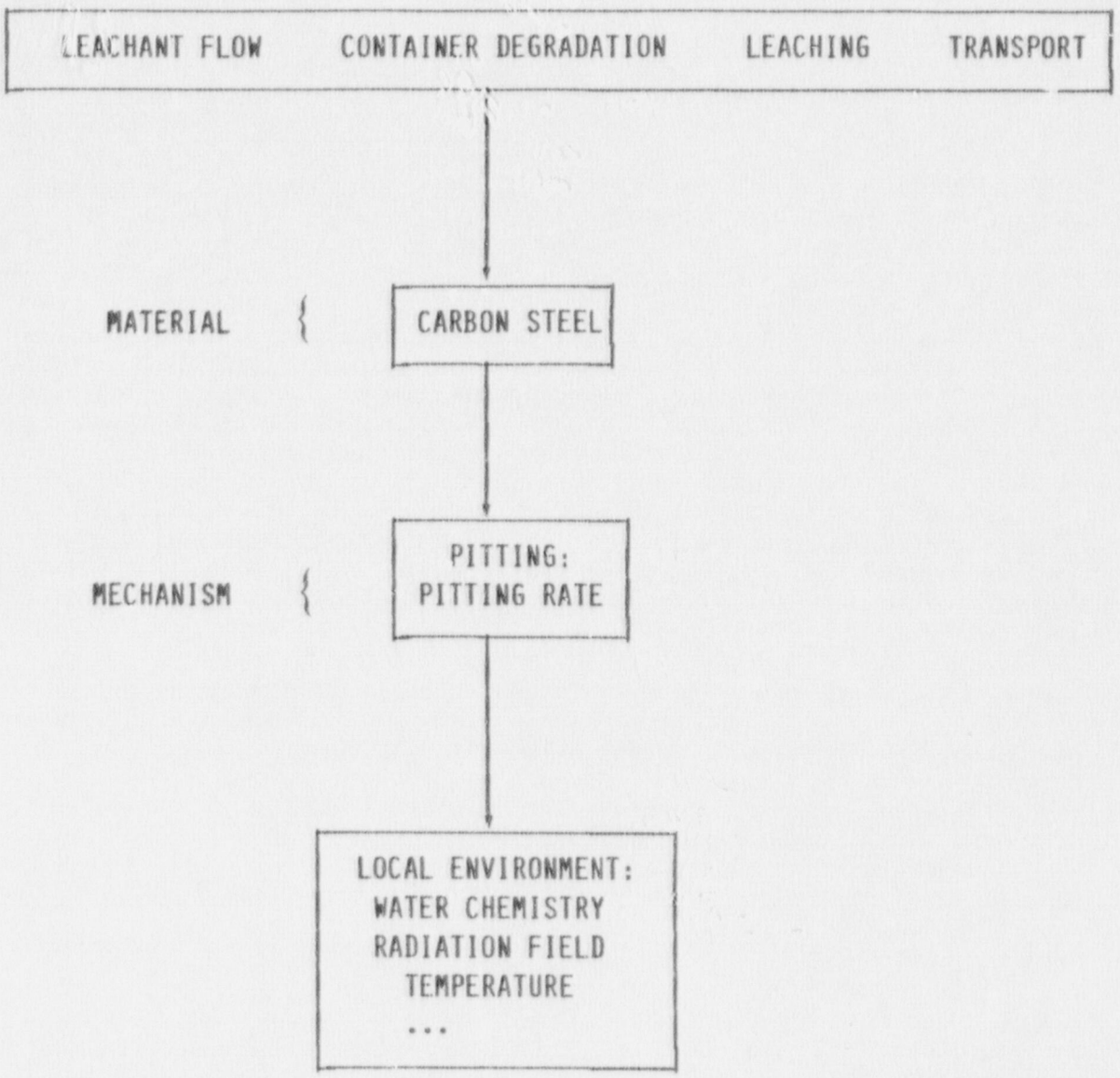


Figure 3.0.2 Schematic conceptulization of one mechanism for container degradation.

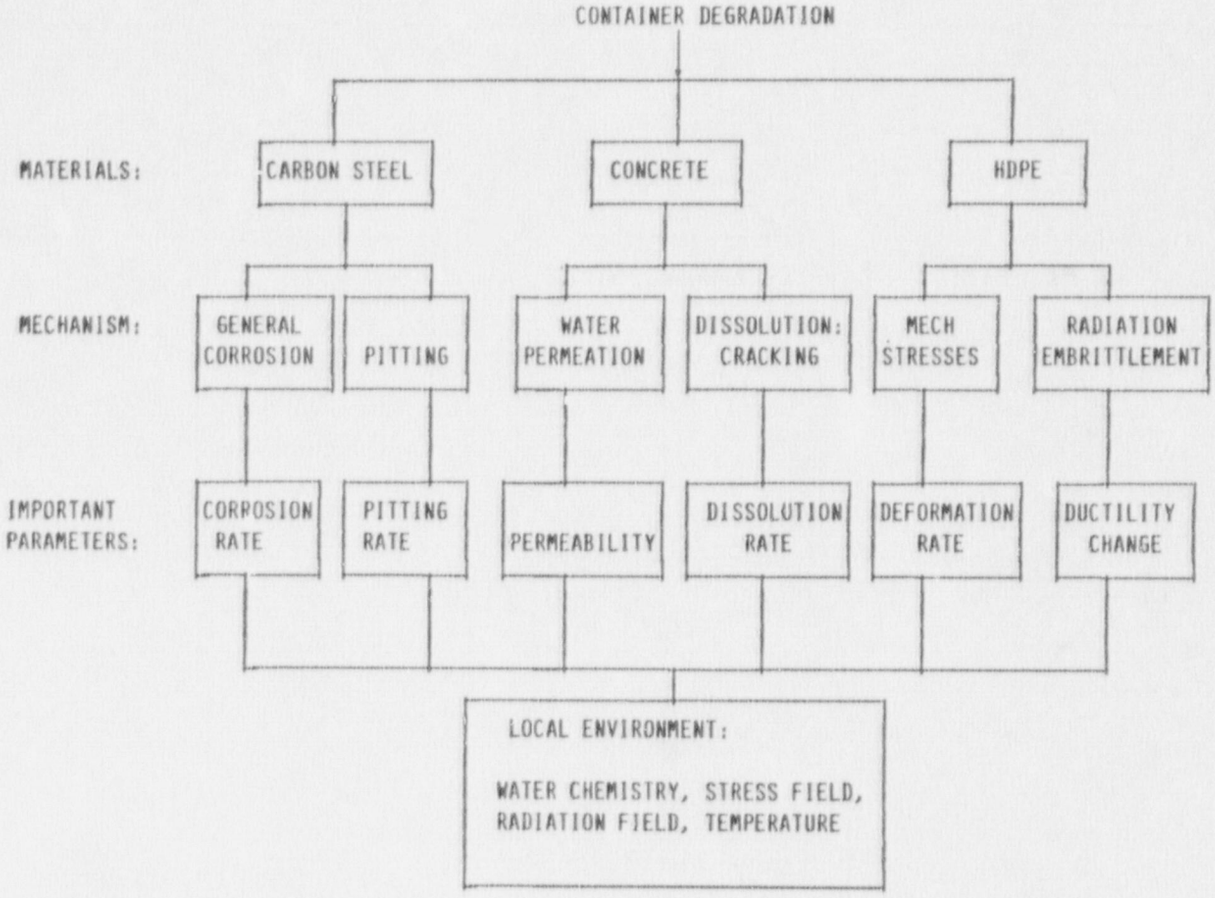


Figure 3.0.3 Schematic conceptualization of container degradation module.

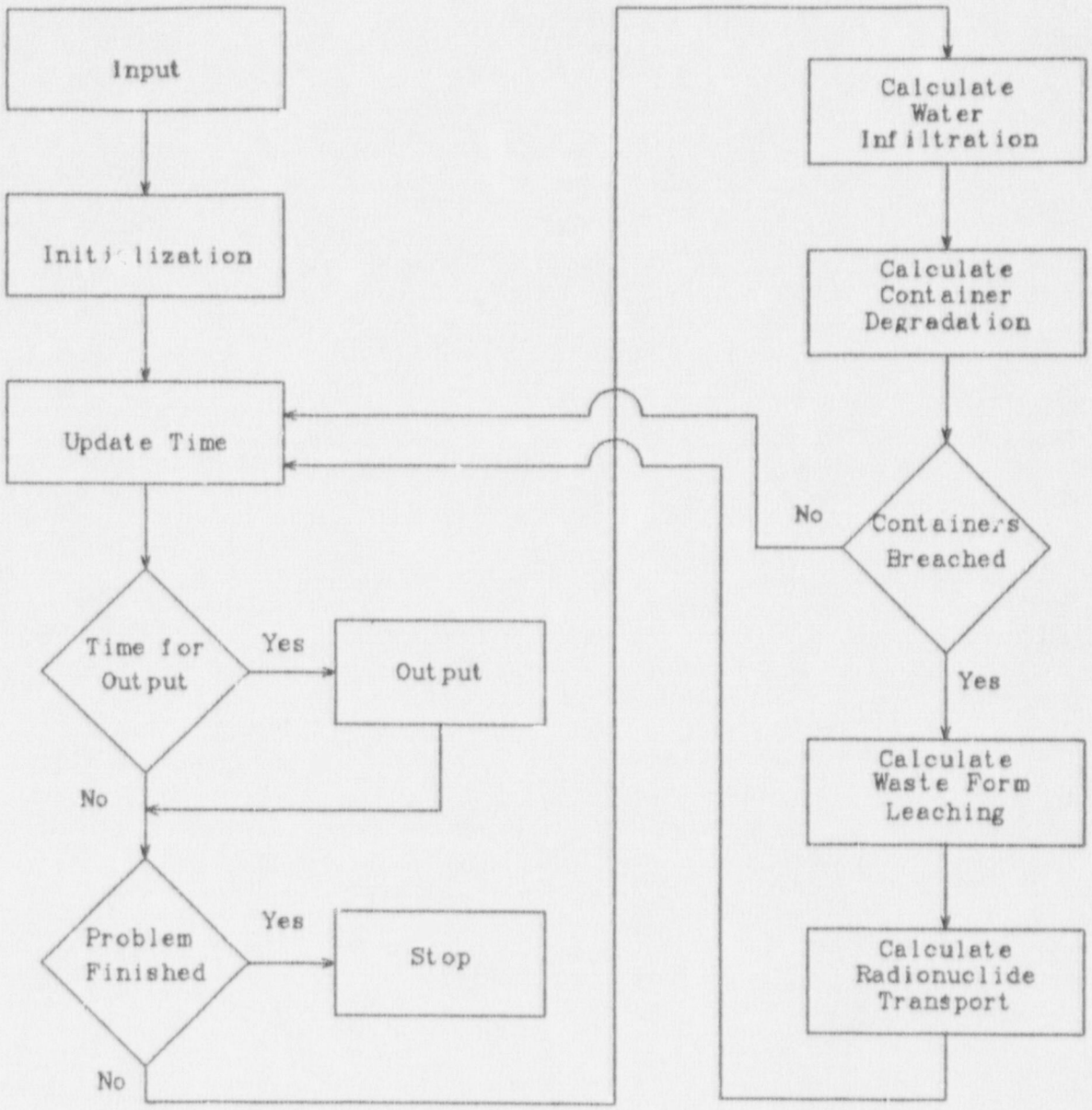


Figure 3.0.4 Flow chart for radionuclide release calculation from a low-level waste disposal site.

3.1 Water Flow Through a Disposal Trench

The most serious problems encountered in shallow land burial are related to water management [General Research Corporation, 1980]. Specifically, for most radionuclides of interest in low-level waste disposal, release from a disposal trench will occur through transport along with water. Further, water will play a major role in the processes leading to release of the radionuclides from the waste form (e.g. leaching, corrosion, dissolution, etc.). Therefore, determination of the quantity and rate of water flow at each location within the trench is critical.

This section presents the basic approach used in modeling water flow in an unsaturated porous medium. The remainder of this section presents a brief discussion of each of the following topics:

Driving forces for water flow;
Sources and sinks of water in the trench;
Influence of trench design on water flow;
Governing equation for water flow; and
Solution techniques for the water flow equation.

3.1.1 Driving Forces for Water Flow

The particular form of the expressions describing behavior depends on the net force operating to drive the process. In general, forces may be expressed as gradients of potential energy. For water in a porous medium, the potential energy can be described as a sum of separate contributions from various factors, e.g., attraction of the soil-solid matrix for water, gravity, osmosis, the action of external gas pressure, etc. [Hillel, 1971]. The total expression for this potential is:

$$H_t = H_g + H_p + H_o + \ldots \text{(other terms)} \qquad (3.1.1)$$

where: H_t = total potential,

H_g = gravitational potential,

H_p = pressure (matric) potential, and

H_o = osmotic potential.

The gravitational potential results from the gravitational field of the earth. The gravitational potential of soil water is determined by the elevation of the water relative to an arbitrary reference level. By convention, this potential is positive if the elevation is above the reference level and

negative below the reference level. At a height z above the reference level, the gravitational potential energy H_g of a mass of water m and volume V is:

$$H_g = mgz = \rho_w gz\ V \qquad (3.1.2)$$

where ρ_w is the density of water and g is the acceleration due to gravity.

The pressure (matric, also called suction) potential arises from the combined action of capillary and adsorption forces on water in the soil. It is a measure of the tendency of the unsaturated porous medium to hold water against suction or a negative pressure force. The source of the capillary action is a result of cohesive and adhesive forces. Cohesion represents the affinity of molecules for their own kind: adhesion is the attraction of ore type of substance for another, e.g., of water molecules and the porous medium. Capillarity in soils occurs through the adhesion of water molecules for particles of soil. Water is regarded as the wetting phase in soil because its soil adhesion tendency exceeds its own molecular cohesive forces. The magnitude of the pressure potential is a function of the pore size distribution, soil type, and volumetric moisture content of the soil. An example of pressure potential versus degree of saturation is presented in Figure 3.1.1. From this figure, it is seen that the pressure potential is zero for a fully saturated soil and negative in unsaturated soils. Greater negative values imply a drier soil with a greater attraction between soil and water.

Osmotic potentials arise from the difference in potential energy between relatively pure water and water containing solutes separated by a semi-permeable membrane. It is not expected that soil particles such as typical alumino-silicate minerals would contain or develop such a membrane. It is more likely that any naturally occurring separation-type structure would be "fully" permeable. Therefore, it is not expected that osmotic phenomena in soils would lead to significant water flow fluctuations and they will be ignored in the modeling of water flow.

There is the possibility that in the pore water of cement waste forms, the concentration of dissolved solids will be much greater than in the surrounding soil water. In this case, the osmotic potential can be an important factor in water flow and will be considered in the equation used to predict moisture migration.

3.1.2 Sources and Sinks for Water

The principal source/sinks for water in the trench system will occur across the boundaries of the trench. At the trench cap/atmosphere boundary, water will be introduced through precipitation and removed through evaporation. At the sides and bottom of the trench, seepage (drainage) into or out of the trench may occur. Another sink for water is uptake by plant roots and eventual transpiration to the atmosphere.

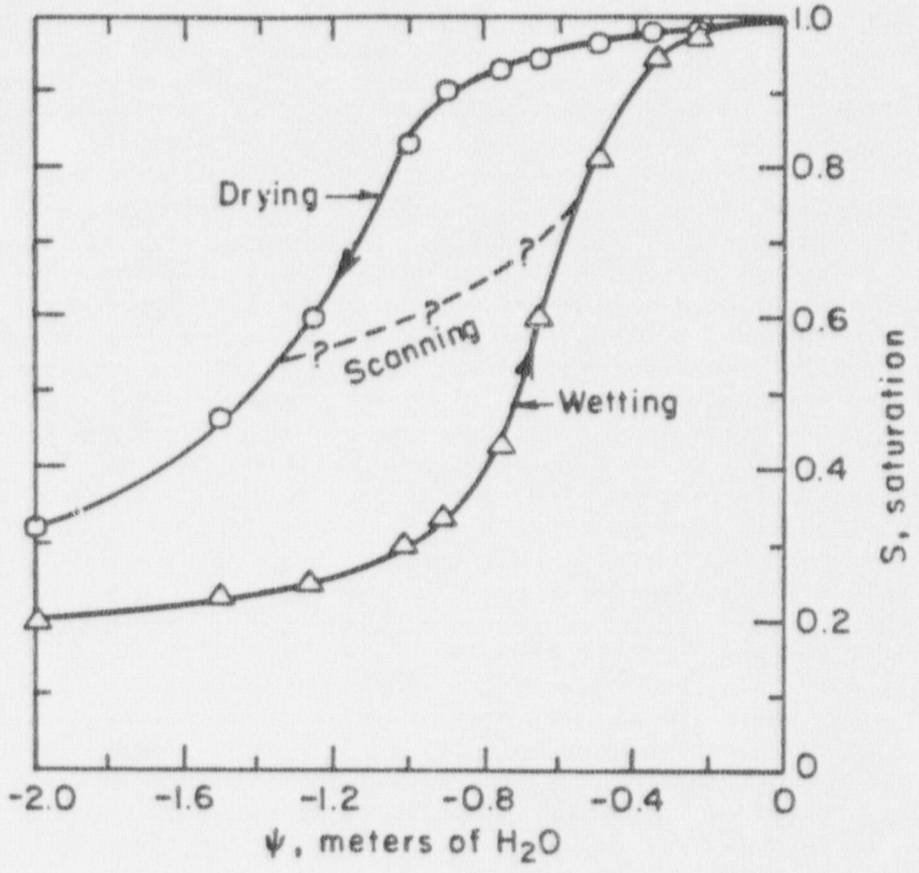


Figure 3.1.1 Example of pressure potential versus degree of saturation (moisture content). Notice the hysteresis between drying and wetting stages (after Liakopoulos, 1965).

In considering the movement of moisture within soils, the starting point is the frequency and amount of precipitation. This precipitation may infiltrate into the soil (trench cap) or depending on the shape of the surface of the trench cap, it may run off the trench cap to the soil surrounding the trench. This should occur to some extent as most trench cap designs call for a sloping surface peaked at the center of the trench. Similarly, if there are depressions in the trench cap due to local subsidence, the water may pond on the surface where it can evaporate or infiltrate the trench cap. For the water that infiltrates the trench cap, a fraction may evaporate at the soil surface, or be taken up by plant roots and stored. Beneath the root zone, water may continue to migrate downward or if the evaporative demands are great enough it may be drawn upward towards the surface.

Evaporation will occur provided that there is excess heat above the latent heat requirements, vapor pressure in the atmosphere is lower than at the soil surface and water exists at the soil surface [Hillel, 1983]. The first two conditions are primarily related to meteorological factors such as air temperature, humidity, wind velocity, and solar radiation. However, strictly speaking, the heat and vapor pressure conditions are not completely independent of the properties of the soils because the energy available for evaporation is a function of the reflectivity, emissivity, and thermal conductivity of the soil. As a first approximation, evaporation is assumed to depend entirely on atmospheric conditions. Similarly, transpiration depends primarily on atmospheric conditions. A common method of treating evapotranspiration is to calculate the maximum amount that could occur given the atmospheric conditions and partitioning the amount transferred by evaporation and transpiration based on the leaf area index, a measure of the ratio of leaf cover to ground area. [Kincaid, 1984]

Drainage out of the bottom of the trench will occur provided the hydraulic conductivity beneath the trench is greater than or equal to the conductivity within the trench. If this is not the case, water will accumulate within the trench giving rise to the "bathtub" effect. The "bathtub" effect has been observed at the disposal site in West Valley, New York.

3.1.3 Influence of Trench Design on Water Flow

The major role of the trench cap is to minimize water flow into the region of the trench that contains the waste. One method of reducing water flow is to use a trench cap composed of different soils. It is well known that a coarse grained material underlying a fine grained material acts as a barrier to infiltration. This process is known as the "wick" effect and results from the fact the hydraulic conductivity for each soil is a strong function of the moisture content of the soil. As water enters the fine grained soil, the hydraulic conductivity exceeds that of the dry coarse grained soil below. Thus, the coarse grained soil acts as a barrier to water flow. To prevent excessive moisture buildup in the upper layer, most trench cap designs have a sloping interface between the two soil layers to produce lateral flow away from the center of the trench.

An example of a trench cap design is presented in Figure 3.1.2. From the figure, the fine grained material is the compacted clay and the coarse grained material is the sand underneath the clay. The gravel layer on top of the clay is used to minimize erosion. Both the sand and gravel layers may act as biological scavangers and provide bio-intrusion controls for the trench cap [Mezga, 1984].

A major concern for any trench cap design is the ability to prevent subsidence. As the waste degrades, consolidation can occur. This can lead to cracking of the trench cap. This leads to a faster pathway for water to the waste and the chance of further collapse as the waste degrades. Modeling of this process for tuff soils and mixtures of bentonite and tuff has been performed [Abeele, 1985].

3.1.4 Equation for Moisture Migration

To fully describe the trench system in terms of water flow, equations of mass conservation are required for the water, air, and soil. Further, an equation for conservation of energy in the system is needed. In the most general case, all of these equations are coupled to each other. For example, water may enter the air phase as water vapor and then its transport would be governed by the movement of the air phase. This is one mechanism for tritium release. Similarly, thermal gradients can lead to enhanced mass flow through bouyancy effects.

Simultaneous solution of the resulting set of coupled differential equations is the most rigorous procedure. However, in many cases the coupling is unimportant and may be neglected. For example, if the temperature changes in the system are unimportant, one can consider the system to be isothermal and thereby eliminate the need for an energy equation. As a first approximation, the energy effects on mass transport will be assumed negligible and the energy equation will not be solved. Also, the soil and waste forms within the trench will be considered immobile, thereby neglecting the need for solution of a "soil" mass balance equation. This ignores the possibility of dissolution/precipitation and redistribution of the soil by the contacting solution.

In developing the numerical formulation for flow in the disposal trench the starting point is the mass balance equations for the water, and air [Oster, 1982]:

air:
$$\frac{\partial}{\partial t}\left[\frac{\phi - \theta}{B_a} + R\,\frac{\theta}{B_w}\right] = \nabla \cdot \frac{K_a(h_a)}{B_a}\,\nabla H_a + \nabla \cdot R\,K_w(h)\,\nabla H_w - S_a \qquad (3.1.3)$$

In this equation the subscript, a, refers to the air phase and, w, to the water phase.

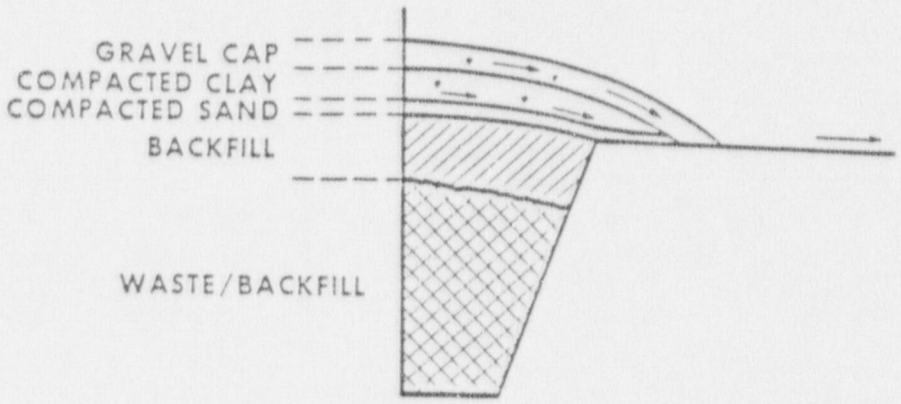


Figure 3.1.2 A conceptual model for trench cap (Skryness, 1982).

Further,

ϕ = porosity,
θ = moisture content,
B_a = media volume factor which is the ratio of volume of air in situ to the volume of air at standard conditions $B_a = 1$ if there is no dissolved air in the water phase.
B_w = the ratio of volume of water with air dissolved in it in situ to the volume of water at standard conditons, $B_w = 1$ if there is no dissolved air,
R = the ratio of the volume of air dissolved in water to the volume of water at standard conditions, $R = 0$ if there is no dissolved air,
$K_i(h_i)$ = conductivity of the ith phase which is a function of the matric potential, h_i
H_i = is the total potential for the ith phase, which is a function of moisture content,
S_i = source/sink term for the ith phase,
h_i = the matric potential for the ith phase which is a function of moisture content.

The mass balance for the air phase states that within a specified volume, the time rate of change of air in this volume is equal to the amount of air transported into the volume plus any source of air in this region. The two terms on the left hand side of Equation (3.1.3) represent the time rate of change of the fractional volume of air in the air phase and air dissolved in the water phase for the volume under consideration. The first two terms on the right hand side represent the net movement of air (including air dissolved in the water phase) into the volume caused by potential flow. The last term represents any sources or sinks of air in this region.

Mass balance for the water phase: $$\frac{\partial}{\partial t}\left[\frac{\theta}{B_w}\right] = \nabla \cdot K_w(h)\ \nabla H_w - S_w \quad (3.1.4)$$

The mass balance, Equation 3.1.4 for the water phase states that the time rate of change of moisture, θ, in a given region equals the amount brought in through flow due to potential gradients plus any sources or sinks in the region. The rate of flow is a function of the hydraulic conductivity. In Equation 3.1.4, moisture movement in the air phase as water vapor is assumed negligible.

To further simplify Equations (3.1.3) and (3.1.4) it is assumed that water flow is much more important than air flow in transport of the contaminants, that there is no dissolved air in the water, and the air is static and remains at atmospheric pressure. For many situations these assumptions are justifiable. In this case, Equation (3.1.3) is no longer needed and the other equation becomes:

$$\frac{\partial \theta}{\partial t} = \nabla \cdot K_w(h) \nabla H_w(\theta) - S_w \qquad (3.1.5)$$

Equation (3.1.5) is the basis for numerical modeling of partially saturated water flow and is known as Richard's equation. In situations where the transport of a contaminant through the air phase is important, such as tritium, both equations, 3.1.3 and 3.1.4, are required along with a mass balance equation for the contaminant (Section 3.3).

In Equation (3.1.3), the change in moisture content is related to the gradient in total hydraulic potential. In order to have the same dependent variable, the variable:

$$C(\theta) = d\theta/dh \qquad (3.1.6)$$

is defined. $C(\theta)$ is the specific moisture capacity of the soil, and h is the matric potential, h_p in Equation 3.1.1. Using the definition, Equation (3.1.5) becomes

$$C(\theta) \frac{\partial h}{\partial t} = \nabla \cdot K_w(h) \nabla h - \nabla \cdot K_w(h) - S_w \qquad (3.1.7)$$

In Equation (3.1.7), the total potential, H, has been divided into the matric potential and gravitational potential. Alternative forms of Equation (3.1.7) in terms of moisture content, θ, have also been developed.

Equation (3.1.7) is the starting point for prediction of the water flow within the trench. Solution of this equation requires:

a) material properties such as the specific moisture capacity and hydraulic conductivity as a function of moisture content;

b) initial conditions such as the initial potential distribution; and

c) boundary conditions which express the amount of water entering and leaving the system as a function of time.

In a disposal trench located above the water table moisture may either enter through the top surface of the trench due to rainfall or may leave through this boundary due to evaporation. Thus, the boundary conditions should be expressed as a function of time. The two simplest boundary conditions for the flow equation are

$$H_w(0,t) = h_0(t) \qquad (3.1.8)$$

and

$$- K(h) \nabla H\Big|_b = q_b(t) \qquad (3.1.9)$$

Where $h_o(t)$ is the potential at the surface and $q_b(t)$ represents the net flux (rainfall minus evapotranspiration) at the soil surface. Similar expressions can be developed for the remaining boundaries of the trench.

At the bottom of the trench, the boundary condition may specify free drainage. In this case, with the assumption that the only potentials are gravity and matric suction, $q_b(t)$ equals K(h) at the boundary and Equation (3.1.9) reduces to:

$$\nabla h\Big|_{x=b} = 0 \qquad (3.1.10)$$

3.1.5 Solution Techniques for the Water Flow Equation

For a realistic description of a disposal trench, Equation (3.1.7) will require numerical solution. Analytical solutions will not be possible because the system is non-homogeneous due to the different materials that will comprise the trench (waste forms, backfill, and trench cap) and this equation is non-linear due to the dependence of hydraulic conductivity and moisture capacity on moisture content.

Various methods exist to reduce the partial differential equation to algebraic equations which can be readily be solved by computational techniques using a computer. These methods include: finite differences, integrated finite differences, finite element, method of characteristics, and random walk methods. The two most common techniques are the finite difference and finite element methods which are described qualitatively below.

In both the finite difference and finite element approaches, the entire system for which the governing equation is to be solved is divided into discrete units for which solutions are approximated. The approximation for each unit results in a matrix representation (a tabular representation of a system of algebraic equations corresponding to the behavior within the unit), and then all of the matrices are coupled together (following application of the appropriate initial and boundary conditions) to obtain the solution for the entire system.

The finite difference method involves division of the system into units of regular geometry while the finite element method handles irregular geometries more easily. The subdivision of the system (here, the low-level waste trench) into a series of units is at the discretion of the modeler. A relatively small number of subdivisions may suffice to give an overall picture

of the water flow behavior through a trench, while increasing the number of subdivisions should lead to a more detailed picture of the water flow. The optimum subdivision will, of course, represent a compromise of the factors of necessary detail (i.e., sufficient to account for water flow around and into waste packages and in the soil), and computational efficiency.

The governing equation for behavior of one aspect of a large system will involve the dependent variable(s) and one or more derivatives of that variable (with respect to time and/or spatial coordinates). The finite difference method involves approximation of the derivatives in the governing differential equation, by a Taylor series expansion which relates the derivatives to the dependent variable. Thus, the partial differential equation can be reduced to an algebraic equation which is a function of the dependent variable only. This process results in a system of algebraic equations for each unit, which, when extended over all the units of the system (the waste trench), amounts to a large number of equations that generally are most easily handled when the equations are given a matrix representation and then solved through matrix manipulation.

The finite element method is a few steps abstracted from the finite difference method. It also involves the subdivision of the system (in this method the units of subdivision are called elements), but it does not continue directly with the estimation of the derivatives in the governing equation differentials by Taylor series expansions. Rather, the finite element approach is based on the realization that any approximate solution to the governing equation has some associated error. One measure of this error is known as the residual and is defined as the difference between the governing equation evaluated using the approximate solution and the governing equation evaluated using the true solution. The finite element method attempts to obtain an approximate solution to the governing equation which minimizes the residual in some manner. Based on principles of variational calculus, the minimization procedure involves multiplication of the governing equation with a weighting function (which is a function of spatial coordinates) and integrating this product over the system volume. The requirement that this integral be zero-valued leads to minimization of the residual with respect to the weighting function. The restated governing equation (called a variational statement) results in a system of algebraic equations, which may then be handled in a manner similar, in principle, to the finite difference approach.

A detailed development of the algebraic equations resulting from applying the finite difference and finite element methods to the water flow equation can be found in a recent progress report [Sullivan, 1986].

A number of codes to calculate water flow in unsaturated porous media have been developed over the last two decades. Compilations of these codes and a description of their strengths and weaknesses can be found in Oster [Oster, 1982] and Kincaid [Kincaid, 1984]. Many of these codes are documented and available to the public through the code authors or the International Ground Water Modeling Center [IGWMC, 1986]. It is likely that one of these codes will be selected for the source term modeling effort and modified to fit the specific needs of this project.

3.2 Container Degradation and Leaching of Wastes - A Model

This section gives the documentation and computational background for a model which has been developed to quantitatively predict radionuclide releases as a function of time from wastes of a porous nature that are disposed of in a corrodible outer container. Information is presented on: 1) wastes to which the model would be applicable; 2) the conceptualization of the processes leading to radionuclide release; 3) the quantitative description of the processes and the calculational flow used to obtain results, coupled with a discussion of the parameters involved in each of the main processes and of the assumptions made in the use of quantitative values of these parameters, along with suggested further refinements and follow-on conceptualized processes for which models are in development; and 4) a series of plots of radionuclide release versus time generated from the model for selected sets of parameters.

3.2.1 Wastes to Which This Model Would Be Applicable

Two broad categories of low-level radioactive wastes may be modeled by an approach such as that presented here. The overlying prerequisites for this model to be applicable are that the waste package consist of an outer corrodible container and an inner porous waste or waste form. (Details of the model are given in the next section.)

The waste categories are:

(1) Class A, B and C wastes that have been solidified in concrete or other porous solid material [assumed in this preliminary stage to be inert and a barrier to radionuclide release by virtue of decreased accessibility to leachant due to tortuosity of the pore space pathways, and by physical obstruction].

(2) Class A non-monolithic heterogeneous lab-trash-type waste in which the radionuclide retention mode is simplified as one of juxtaposition, not involving adsorption or chemical binding to the trash material (i.e., paper, rubber gloves, etc.).

3.2.2 Conceptualization of the Processes Leading to Radionuclide Release

In descriptive overview, the model for radionuclide release from these wastes may be conceptualized as summarized below. Figure 3.2.1 gives the sequences of events and processes as they relate to each other over time. The capital letters in the figure correspond to the descriptive paragraphs given here.

A. The first input to the model is the water flow. This water influx must be specified as a function of time, and x and y coordinates, such that it is physically localized at the waste package "top" surface. In the model, water flow is given the symbol, W, with units of length$\cdot$time^{-1}, i.e., representing a column of water of unit area passing in a time interval.

B. An induction period may be given in which the containers have begun corrosion but have not yet been breached by a pit (given the symbol, t_o, with units of time). The length of this period will vary with site-, and waste-package-specific characteristics.

C. Initial pit formation occurs in the upper portion of the waste container such that water influx through the pit is physically restricted by the size of the pit. The pit is assumed to be circular and to grow larger with time until a limiting size is reached. Once this has occurred, physical restriction of water influx is no longer dependent on time (the pit area is given the symbol, P, and has units of length^2).

D. Water flow occurs through the pit in the outer container. This is assumed to proceed with time (the pit area is growing as well) until the available pore or void space of the waste package (and, for monolithic wastes, of the waste "form" itself) is filled (the available pore or void space of the waste "form" is given the symbol V and has the units length^3).

E. Second pit formation occurs; this can serve as an exit for leachate. This process is assumed to take place at a time equivalent to an induction period following the first pit formation. For example, if the induction period to first pitting is taken as five years, the second pit will be assumed to occur at ten years. It should be noted that the first pit is assumed to grow continuously during the intervening period before second pitting (and afterwards, as well).

F. Once the pore and void spaces of the waste are occupied by water, the process of exit of this water (now essentially, leachate) may begin. The increment of water taken "into" the container is, for that time interval, to be a replacement for an equivalent volume of water exiting the waste laden with "leached" radionuclides. The release of radionuclides from the waste to the water may be thought of as consisting of four phases (which may overlap chronologically) as follows:

(1) removal of outermost surface species by a surface "wash-off",

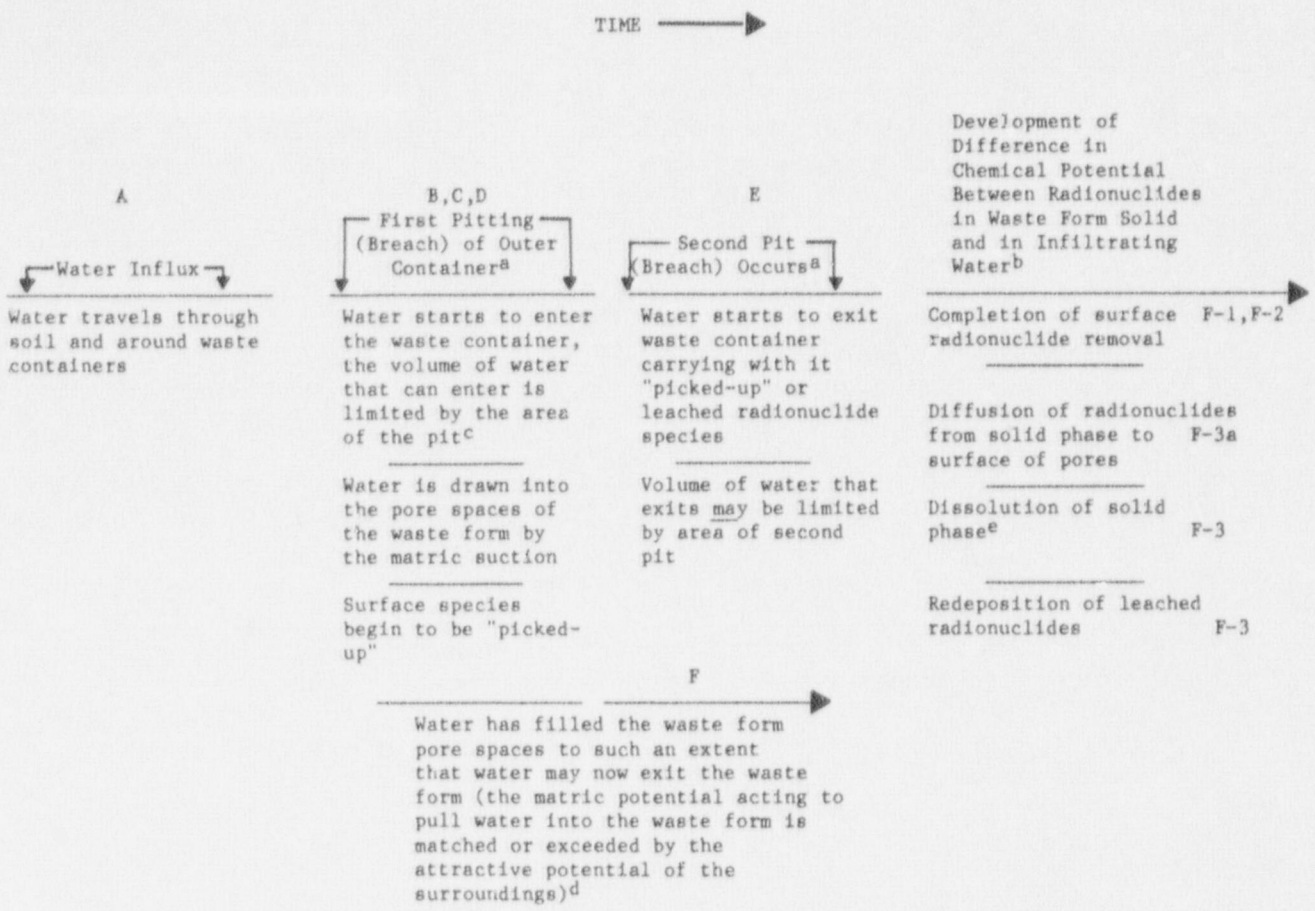
(2) removal of species residing on the inner pore surfaces,

(3) removal of radionuclides incorporated in the waste matrix or solid by:

a) diffusion through the waste solid matrix to the pore space surface and subsequent "pick-up" by leachant,

and/or,

b) dissolution of the matrix material such that further intrusions are produced, which lend access for leachant to radionuclide species,



[a] A range of time for pitting has been given due to known variability in materials, environments, and in corrosion itself for a given environment and material; pitting corrosion is just one type of corrosion that might occur.

[b] A difference in chemical potential between radionuclides residing in the bulk solid part of the waste form and those dissolved in the infiltrating water would theoretically begin as soon as the first radionuclides were "picked-up" from the surfaces of pores. Diffusion from the solid phase would be expected to commence, driven by the difference in chemical potential. A range of time for the development of this difference in chemical potentials is given because it is dependent on the progress of leaching which depends on water flow and on any restrictions to water flow (such as outer container breach).

[c] Limitation of water influx into waste container by the outer pit size can be terminated at a number of points; two choices that have been modeled are: 1) corrosion pit size reaches an area equivalent to a drum top, and 2) corrosion total area reaches the equivalent of 50% of the total drum area.

[d] An assumption implicit in this is that "net" or "bulk" water flow is slow relative to its uptake into porous media due to matric potential.

[e] Solid dissolution may actually begin from the first contact with water. However, this is believed to be quite slow and has been neglected at early times in the first modeling phase.

Figure 3.2.1 Chronological representation of main processes occurring in leaching model.

and,

(4) redeposition of radionuclides along the waste pore surfaces by adsorption, or through plugging of the pore spaces (this may occur by carbonation or other mechanisms). This represents competition of solid phase materials with leachant for solute radionuclides.

G. Leachate "laden" with leached radionuclides exits the waste package. This process may be complicated by competition of solid phase materials encountered along the leachant's exit pathway for radionuclides contained in the water (e.g., sorption sites may become available in drying cycles so that "picked-up" species re-deposit in the waste or on the outer container).

The model being presented here incorporates processes A through F(2).

3.2.2.1 Interactions of Waste with Matrix and Water

Waste radionuclides in porous wastes (and solidified waste forms) may be thought of as existing in two main physical "states". These are:

i) radionuclides residing at the outer surface of the waste form and/or at the surface of pores inside the waste form; these have been referred to as "surface species" or "surface radionuclides" [these are participants in processes F(1) and F(2)], and,

ii) radionuclides residing "within" the solid fraction of the matrix material itself. These radionuclides may come in contact with leachant through two processes:

a) either the water reaches the radionuclide (through diffusion, crack formation), or,

b) the radionuclide diffuses through the solid matrix to the pore water [these two processes correspond to F(3)].

Of the two major "states" of radionuclides just given, it would be expected that the more easily accessed, and hence, removed, would be the "surface species." For radionuclides in chemical states that are relatively simple, e.g., hydrated cations, the retention at the surface and in the solid matrix may be one of simple spatial accommodation (with, possibly, a van der Waals-type component). In such a case a reasonable model for the "pick-up" of radionuclides by leachant would be a "contact"-type leaching, i.e., the radionuclide would move into the water on contact (given that solubility limits are not exceeded).

For radionuclides whose retention mode is by some form of chemical bonding (adsorption/ionic attraction/adherence to an ion exchange resin) the movement into the leachant may still take place but at a slower rate. This "pick-up" could be modeled as a two-step process:

1) contact of leachant with the region of radionuclide deposition,

and,

2) "exchange" of radionuclide between the solid phase and the leachant with exchange rate constants, k_e and k_e', expressible as

$$[\text{Radionuclide}]_{\text{solid}} \underset{k_{e'}}{\overset{k_e}{\rightleftharpoons}} [\text{Radionuclide}]_{\text{leachate}}$$

The value of k_e and, similarly, of the rate constant for the reverse reaction, k'_e will, of course, depend on a large number of factors (e.g., the particular chemistry of the solid phase and of the radionuclide, the mode or type of adherence to the solid phase compared to the type of situation available to the radionuclide in the leachant environment, etc.) A mode of simple physical accommodation of the waste radionuclides by the solid material is being assumed in the model presented here.

3.2.2.2 Calculation of Surface Species "Source"

The calculation of the surface species "source" has been performed on the basis of two assumptions:

1. in production of the pores and void spaces for a monolithic waste, the deposition of the radionuclides originally in the pore or voids occurs at the surface of the pore or void (in other words, the pore formation is seen to represent a folding in or collapse of material in which the radionuclides were originally homogeneously distributed), and,

2. the pore surfaces still contain their original concentration of radionuclides such as would have been present prior to the folding in of the pore spaces.

The first assumption leads to a straightforward estimation of the surface species "source", namely, a 10% pore/void space waste would have 10% surface species, 20% pore/void space would lead to 20% surface species, etc.

A correction to this occurs by virtue of assumption 2. There will, in essence, be >10% surface species in a 10% pore/void space waste because there were radionuclides originally throughout the waste, the production of the pore spaces is assumed not to lead to re-distribution of the radionuclides throughout the remainder of the waste (i.e., 10% pore/void production is assumed not to lead to redistribution in the 90% solid fraction).

The determination of the contribution from assumption 2 has been based on a conceptualization of a solid cube composed of spheres of equivalent size. A percentage of the spheres are species A, while 100%-A% are B. The analogy to wastes and radionuclides is that A may represent radionuclides while B may represent waste form or solid fraction material. The assumption of a homogeneous distribution of A throughout the cube leads to a certain percentage of A on the surface. Using geometric arguments it can be shown that the number of surface spheres is:

$$S = n^3 - (n-2)^3$$

where S = number of surface spheres, and
n = number of spheres per edge.

For example:

1. cube 1, 1000 spheres, 10 per edge

   512 spheres are "inside" and 488 are on the outer surface. If the cube had a 10% A fraction, then 51 A spheres would be inside the cube and 49 would be on the outer surface. This means 51% of the A sphere fraction would be inside with 49% outside. This cube has a 10:1 ratio of cube edge size to sphere size.

2. cube 2, 10^6 spheres, 100 per edge

   9.4×10^5 spheres are "inside" and 5.9×10^4 are on the outer surface. If the cube had a 10% A fraction, 5.9×10^3 A spheres would be on the surface of the cube and 9.4×10^4 spheres would be "inside" the cube. The surface fraction of A spheres would be $5.9 \times 10^3/(5.9 \times 10^3 + 9.4 \times 10^4) = 6\%$. This cube has a 100:1 ratio of cube edge size to sphere size.

As can be seen from these two examples, the surface fraction is highly dependent on the relative sizes of the "object" (here, the cube) and the radionuclide (here, sphere A).

Capillary pore and gel pore sizes for concretes have been reported [Neville, 1981] to be 1.3×10^{-4}cm and 2×10^{-7}cm in diameter, respectively. The smaller of these two, the gel pore diameter, represents approximately 10 times the order of magnitude size for atomic diameters, while the larger is $\approx 10^4$ times atomic diameter size. A conservative approach has been taken that pore space diameters (analogous to cube edges) are $\approx$100 times the atomic diameter size. This would lead to an approximation of a cube edge 100x the size of an individual sphere (a 100:1 ratio, as in example 2). Given the sphere/cube conceptualization discussed earlier, an idealized solid waste form composed of a great number of "cubes" whose edges joined to form the pores, would lead to a 100:1 solid fraction size to radionuclide size ratio. From this, the 6% surface species fraction may be inferred. For the purpose

of calculation in this model, this has been rounded to 5% and serves as the 5% addition to the pore fraction percentage seen at the top in Figure 3.2.5 where the calculation of the pore surface species fraction has been outlined.

3.2.2.3 Model Conceptualization Summary and Assumptions

In short, water is taken in to the waste through a corroded opening in the outer container; the water continues to enter the pore and void spaces (i.e., space in which capillary and adsorption sites are available and which, therefore exerts a "matric" suction [Kempf, 1986]) until the pore and void spaces are filled; then the surface species are removed on the basis of a "rinsing" process (similar, in principle, to that developed in Kempf, 1983). In this process, the waste pore spaces filled with water are considered similar to a volume of solution to which new water volumes are added sequentially (this represents rain-water influx periods) and from which, coincident with the influx, equivalent volumes of leachate are allowed to exit. The waste form is assumed to maintain a steady state as far as total volume of water contained (it is assumed that the change in volume due to removal of pore surface species radionuclides is insignificant at this stage; in other words, solid dissolution is ignored).

Implicit in this model conceptualization is that the pore spaces of the waste are connected. Figure 3.2.2(a) is an idealized sketch of the outer edges of two porous wastes in cross-section. Figure 3.2.2(a)(i) represents a waste in which the pores are connected, and Figure 3.2.2(a)(ii) represents a waste with non-connected pore spaces. This section treats wastes similar to Figure 3.2.2(a)(i). To model leaching from a waste similar to that in Figure 3.2.2(a)(ii), the approach could be used that a certain volume percent of pore spaces, X, were interconnected and could be treated by this model, while the 100-X% remaining were participating in a leaching mode containing a diffusion term (either radionuclide diffusion through solid to "connected" pore space water, or water diffusion through solid to "non-connected" pore space radionuclides, or both) which would introduce a release term chronologically delayed compared to the leaching modeled for Figure 3.2.2(a)(i).

Figure 3.2.2(b) illustrates this model as time increases (the sketches are not meant to be spaced over equal time intervals). The first sketch represents a waste container on which water is incident and then allowed simply to flow over the container edge back to the surroundings. In (ii) a corrosion pit has occurred in the container top, thus allowing some water influx. The shaded area represents that volume of the waste for which the pore and void spaces are being filled with incoming water. In (iii), the first pit has grown in area, thus allowing more water in, and also, a second pit has formed in the bottom of the container. The placement of the pits has been chosen to reflect those conditions which are expected to be reasonably realistic but also to lead to the most effective leaching of the waste (and therefore to yield "conservative" results, see Kempf, 1986). The water-filled pore space has correspondingly increased in volume. In (iv), the first and second pits have grown in area and the pore and void spaces of the waste have been saturated. The next increment of water taken in is expected to "push" an equivalent increment of leachate out, as shown in (v). The process occurring

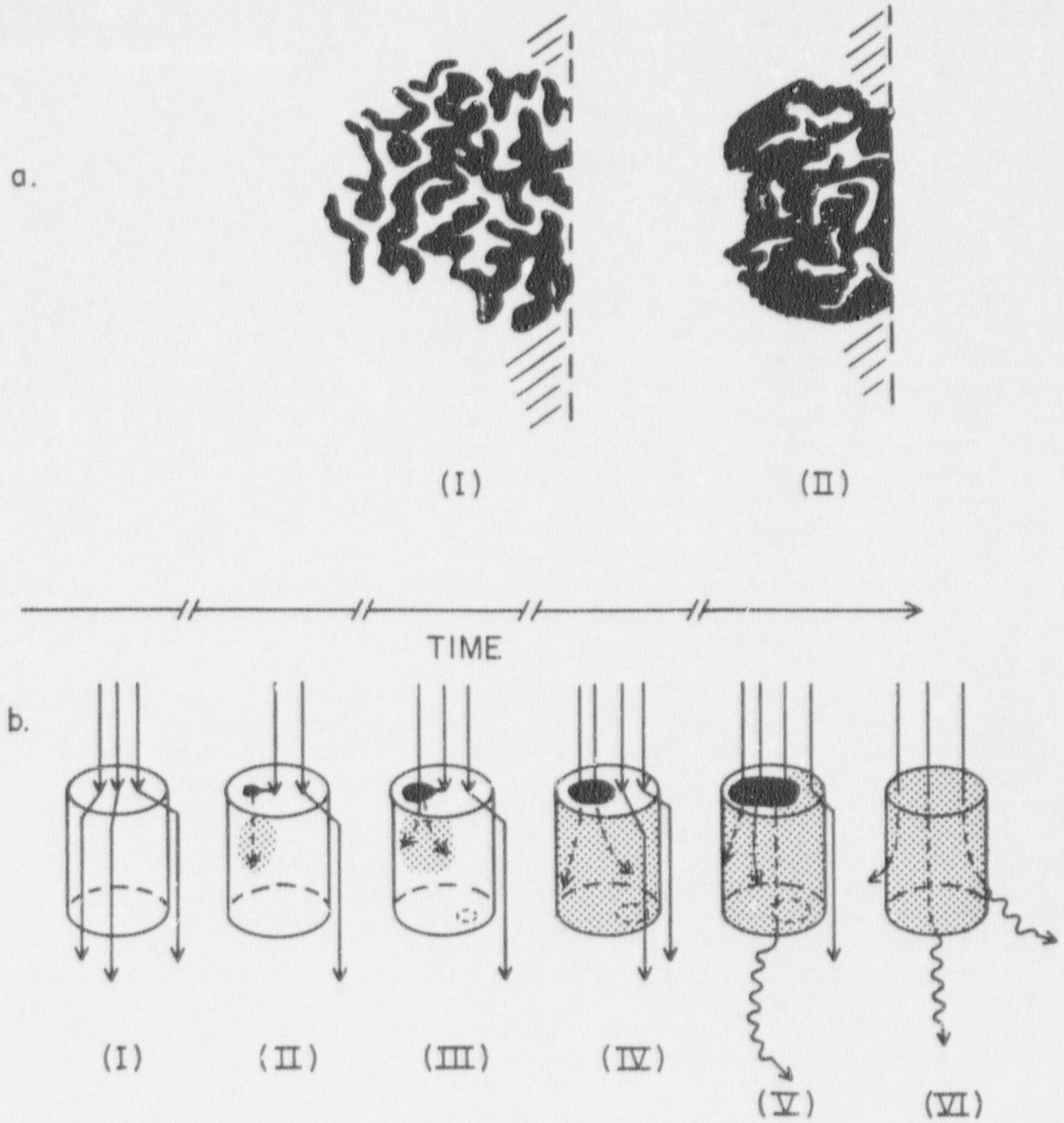


Figure 3.2.2 (a) Cross-sections of idealized wastes with (i) pores connected, and (ii) pores not connected; (b) sequence of main processes in leaching model for contained porous wastes.

in (v) is modeled to continue until the pore surface species "source" is depleted. Figure 3.2.2(b)(vi) represents a monolithic waste form for which the outer container has been totally breached or which can no longer be expected to restrict leaching. In such a case, the incident water may infiltrate at a greater number of locations and, similarly, exit of leachate may occur at a number of locations.

It is understood that the outer container in which wastes are packaged may be of a type for which the principal failure mode is expected to be corrosion; in this discussion of model development, carbon steel is assumed to be the material of interest, but the model itself could be applied to any corrodible material, given that the appropriate parameters in the model were customized to the specific behavior of that material. For example, breach of a stainless steel may be modeled to occur by a pitting mechanism, but the rate constant for such corrosion would need to be that applicable to the particular stainless steel in the given environment.

3.2.3 Quantitative Description of Processes and Calculational Flow Used to Obtain Results

The quantitative factors (aside from time) involved in this model are:

(a) water influx amount, W_i,
(b) pit area, P_i
(c) incremental "rinse" volume, ℓ_i, and cumulative "rinse" volume, V,
(d) surface species "source", $(X_o)_i$, and
(e) incremental and cumulative amounts of radionuclides released, IFR, and CFR.

3.2.3.1 Water Flow

The water flow used in this model is expected to be generated from a water flow code. For the purposes of obtaining quantitative results for this segment of the entire waste site/waste form leaching system, a series of water flow values, W_i, have been assumed. In particular, for the data sets generated with this model, values of 1, 10, and 100 (units of length/units time) have been assumed (cm and year have been adopted for these calculations, although any length and time units could be used so long as they were maintained consistently with other parameters in the model). Earlier work on models for radionuclide release from waste packages from the Sheffield burial site used water flow values corresponding to the rainfall at the ground surface of the burial site (89 cm/year); rainfall that reaches the water table at the site (6.35 cm/year) and the average of these two extremes (48.3 cm/year) [Kempf, 1983]. It is felt that the range of values covered by letting W_i take on the magnitudes 1, 10, and 100 cm/year represents a realistic range of amounts of water that may be seen by the low-level wastes at a burial site.

Net water flow will be downward because neighboring sections will also be receiving incident flow. Lateral matric potentials will not be experienced by

vertical component flow because they will be diminished or exhausted by the flow in the adjacent sections.

In the limit of soil column sizes, with the incident rainfall filling soil pore spaces and thus eliminating matric potential, the net matric potential becomes unidirectional (z component).

This can be expressed by:

$$\lim_{\substack{V_{sc} \to 0 \\ V_{spsu} \to 0}} \phi_M(\text{total}) = \phi_M(\text{z direction})$$

where ϕ_M is the matric potential,

(total) corresponds to x, y, and z components,

z direction corresponds to vertical (downward) flow,

V_{sc} is the void and pore space volume for a soil column,

V_{spsu} is the volume in a soil column corresponding to unoccupied pore space.

In other words, in the limit as the available soil column void and pore space approaches zero, the lateral matric potentials from adjacent soil columns tend to approach zero and the vertical or z-direction potential (matric and gravitational) is all that remains. Figure 3.2.3(a) shows a schematic version of soil units divided into soil columns. The incident water flow is illustrated as the vertical arrows and it can be visualized from this sketch that, given evenly distributed incident water, the lateral tendency for flow will be minimal.

This is significant and applicable only until the homogeneity of the solid is disturbed, e.g., a layer of waste containers surrounded by backfill will constitute a disturbance such that a limiting approximation of soil column size will be invalid. However, backfill between containers may be approximated in this manner, up to within a certain spatial limit at the periphery of a waste container. [See Figures 3.2.3(b) and 3.2.3(c).]

Water flow downward through the soil until contact is made with a waste container may be simplified as having two possible results [as illustrated in Figure 3.2.3(d)]. First, the water strikes the waste container surface, flows along its top until it reaches the edge and then resumes downward flow (it is understood that some "pooling" of water may be necessary before this flow across the container top can occur, but this is likely to involve a small amount of water and the net effect will be the same), and second, the water reaches the upper surface of the waste container and, in its flow along the surface, encounters a breach and enters through the breach. This has been taken as the basis of the model as discussed in Section 3.2.2.

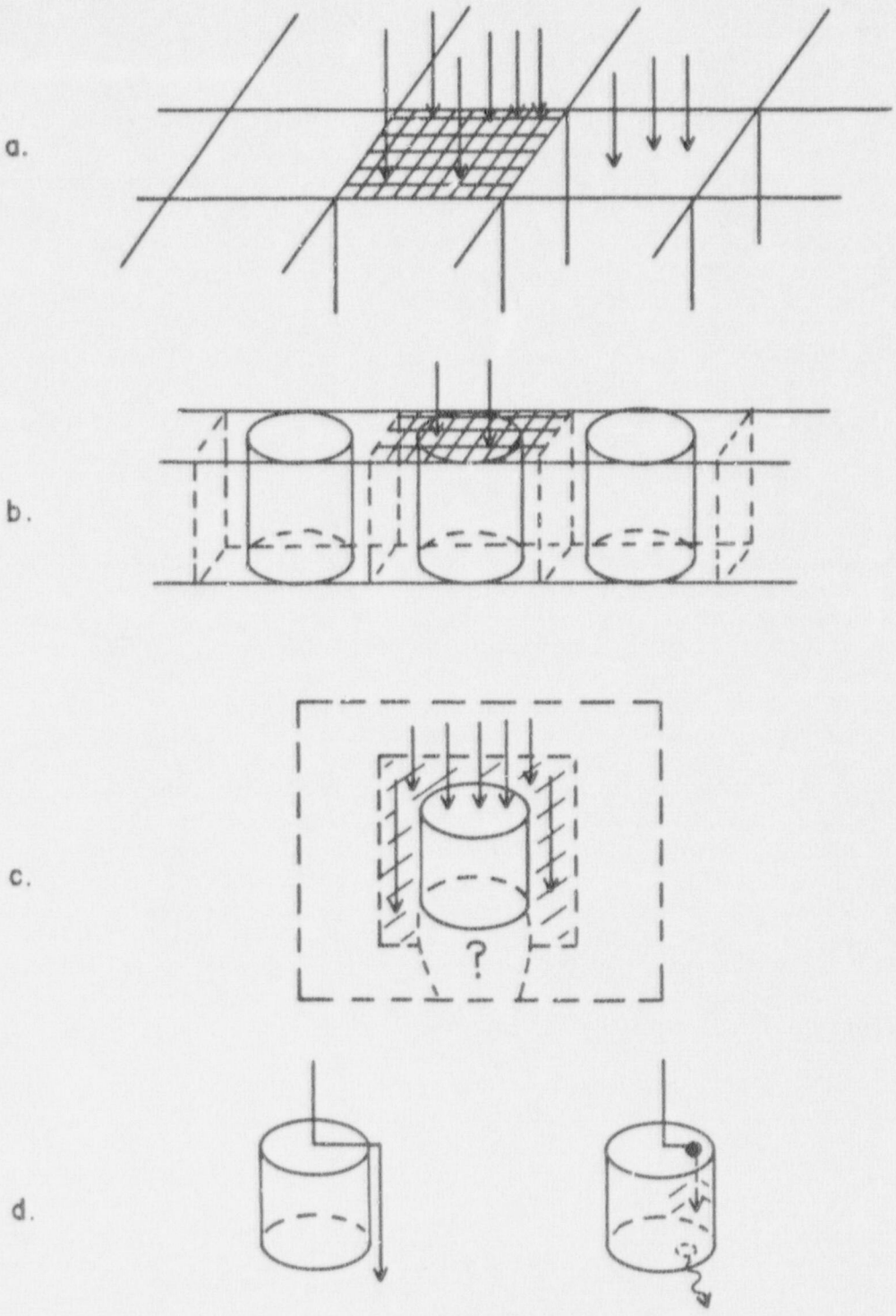


Figure 3.2.3 (a) water flow through soil columns,
(b) influence of waste containers on soil column size assignment,
(c) pattern of flow in vicinity of waste container,
(d) water flow in leaching model.

3.2.3.2 Outer Container Corrosion

The conceptualization of restricted water flow through a corroded area of the outer container leads to a need for a quantitative expression for the corrosion rate. The total pit area is what limits the water flow in this model and, for simplicity, circular pit growth has been assumed. Should more than one pit actually occur, the model could accommodate this through the appropriate setting of the value for the corroded area. For a circular pit, the growth rate of the radius may be expressed as:

$$r = kt^n$$

where

r = pit radius
k = corrosion rate constant (site-, and material-specific),
t = time
n = exponent for dependence of the corrosion rate on time.

In this model, time has been set as an independent variable while a range of values have been taken for both k and n. The rate constant, k, has been given the values 0.95 cm/year and 0.095 cm/year. The former value corresponds to a pit depth rate observed for carbon steel materials in a Sheffield-type (humid) environment [Romanoff, 1957; MacKenzie, 1985], while the other end of the range, k=0.095 cm/year, has been arbitrarily assumed, to represent an environment much less conducive to corrosion of carbon steel (such as what may be experienced in a more arid environment). It should be noted that calculations of total drum lifetime with this k value and a square root time dependence lead to extremely long drum lifetimes. This is considered unrealistic and it must be reiterated that carbon steel drums in a soil environment should not have lifetimes exceeding 120-150 years [MacKenzie, 1985]. Use has been made of a pit depth rate value for a parameter which represents areal pit growth (with an assumed depth equivalent to the carbon steel thickness). This implies an aspect ratio of one, which is felt to be reasonable for carbon steel. The time dependence, n, has been given the values 1 and 1/2; this should give a range of results, i.e., a rapid corrosion rate, and a relatively slow corrosion rate.

The circular pit area may be calculated by substitution of the expression for the radius, r, into the formula for the area of the circle:

$$P = \pi r^2$$

$$= \pi(kt^n)^2$$

$$= \pi\, k^2 t^{2n}$$

Thus, from this expression the area of corrosion in the outer container can be calculated for any value of t. As mentioned in Section 3.2.2, an induction period, t_o, is expected to pass before pitting begins. This t_o has a value that is highly dependent on material and external conditions. It will vary from site to site. For applications of this model, total times to

pitting, release, etc. have been calculated as starting at the end of the pertinent induction period, t_o. This was done to make the results generated as general as possible.

As a simplification to the model, it is assumed that in the time required to accumulate sufficient infiltrating water to saturate the waste pore space volume, another (or more than one) pit has formed in the outer container so that leachate may exit the package. This means that the induction period to second pitting is exceeded by the water accumulation time. There may very well be instances when this is not the case; these would be situations in which the accumulated water in the waste drum would need to "wait" for exit. In such an instance, it might be expected that leachate exit could be restricted by the second pit size. Second pitting prior to total pore space saturation is considered a reasonable assumption; also the most conservative case of second (or greater than second) pitting at the bottom of the container has been taken to maximize expected release (at this point, it is not certain that second pitting at other locations on the drum would significantly inhibit release; it may simply be a case of a further lag time of some years, until a "pool" of water had accumulated to the height of the next pit before release, or exit, could occur).

The pit size could, theoretically, be allowed to continue growth until the entire area of the outer container had corroded away. This is considered unrealistic from the point of view of the significance of restriction of water influx based on the pit area. Thus, two choices (out of an infinite number of possibilities) have been chosen as upper limits of the water influx area. They are:

a) corroded area equivalent to the drum top area,

and

b) corroded area equivalent to 50% of the total drum area.

For the calculations whose results are presented as a part of this section, the first choice has been assumed, i.e., the pit area was allowed to grow with increasing time until the total became > the drum top area. At this time (and thereafter), the water influx was taken as limited by an opening the size of the drum top area. References to the water influx, corroded pit area, and limiting influx area are made as a part of the calculational flow chart given in Figure 3.2.4. (Further discussions of incremental influx, pore volume accumulation and surface species leaching will refer to this figure and to Figure 3.2.5, as well.) A dashed line has been used in Figure 3.2.4 to connect the calculation flow points surrounding choice b. Were choice b made, these lines would indicate the process to be used for calculation.

3.2.3.3 "Rinse" Volumes - Water Influx to the Waste Itself

Leaching of containerized waste forms can be expected to begin once a breach has occurred in the outer container such that moisture can enter and contact the waste form. The processes that occur on this contact will depend

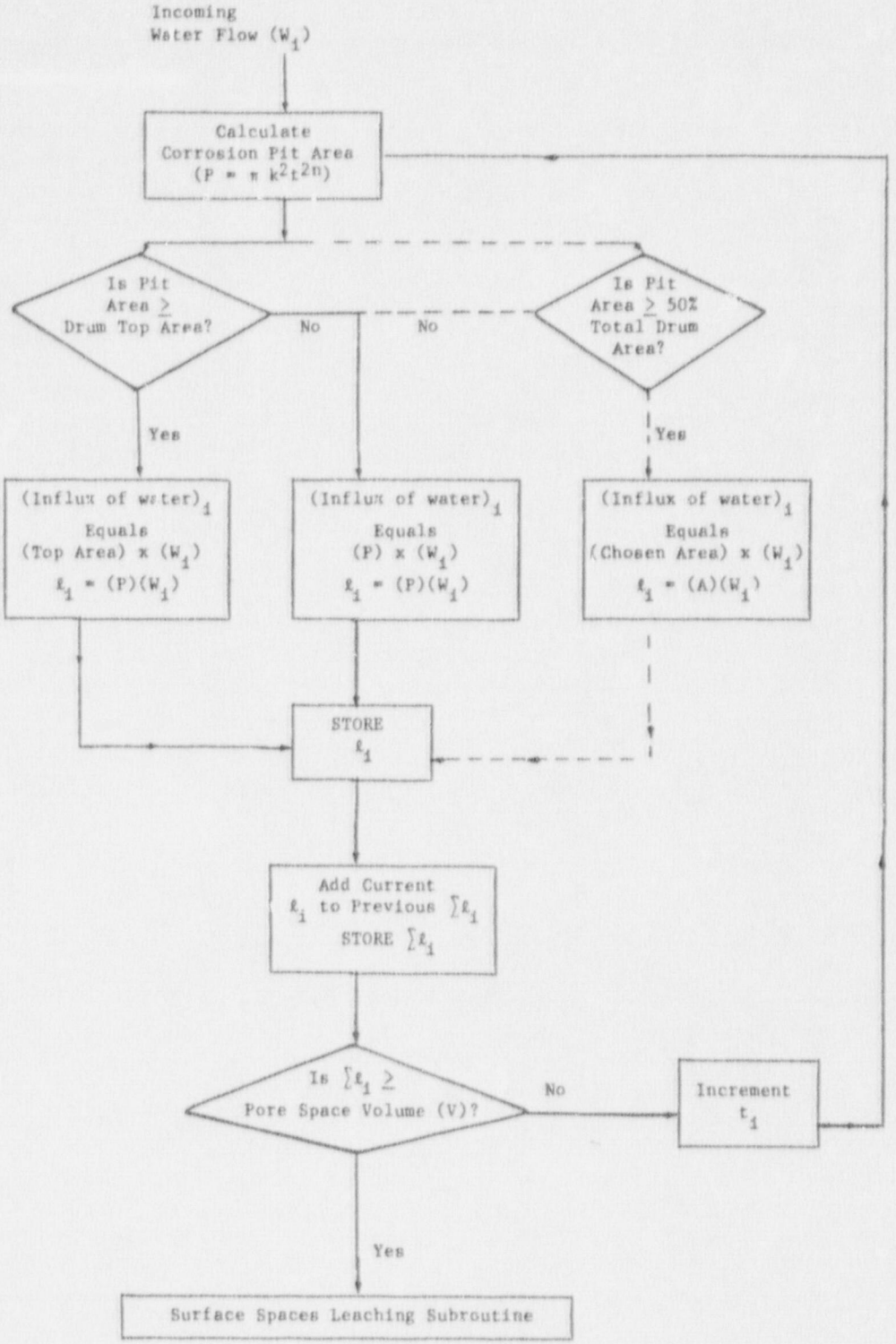


Figure 3.2.4 Calculation flow for radionuclide release from porous wastes in a corrodible outer container.

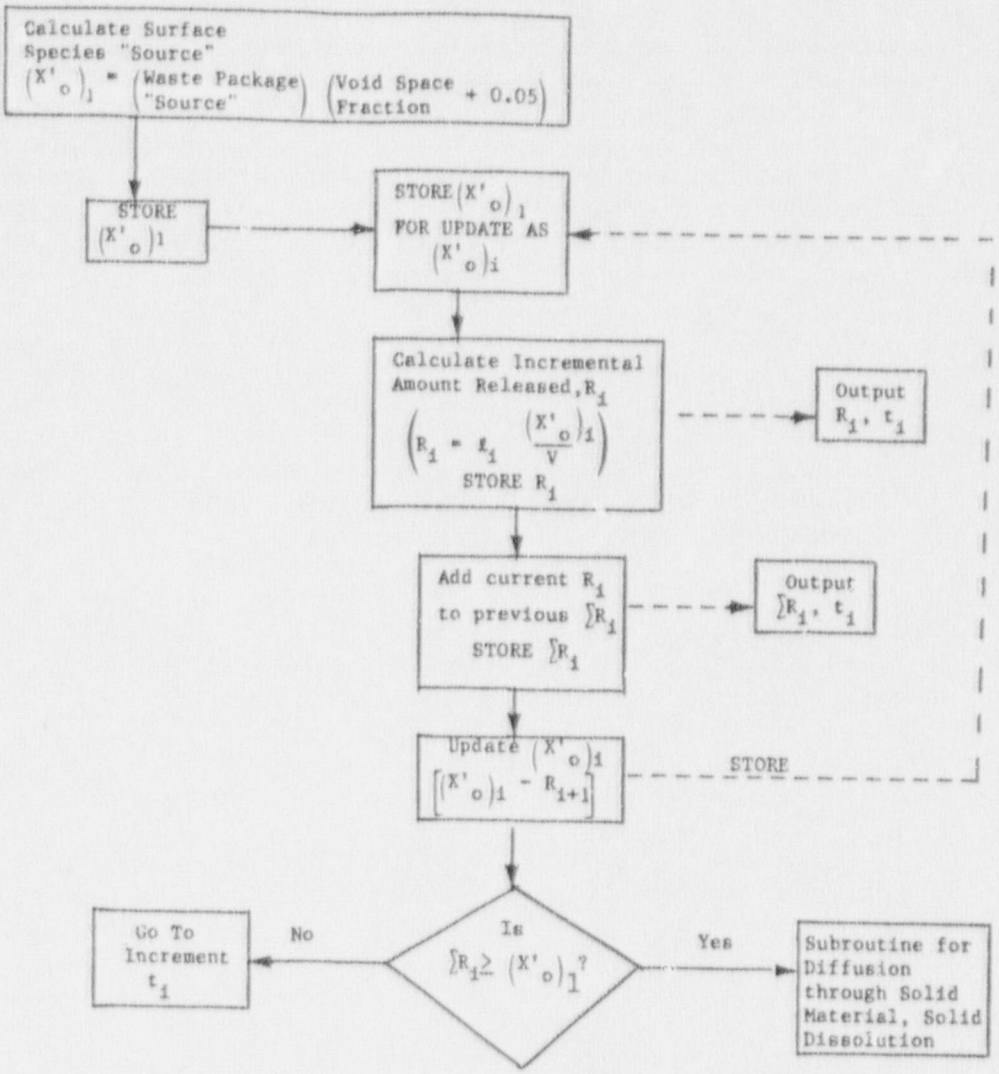


Figure 3.2.5 Surface species leaching subroutine.

on whether the waste is porous (e.g., concrete-solidified wastes) or encapsulating (e.g., bitumenized wastes). For porous waste forms, on which this discussion is centered, infiltrating water will tend to be "pulled" in by the matric potential of the porous medium (this "matric" potential, resulting from availability of capillary and adsorption sites, has been discussed previously, Kempf, 1986). The magnitude of this matric potential will vary with the type of porous medium; for concrete, which is used as a solidification agent for much low-level waste, the matric potential will vary with (among other things) the amount of pore water already in the concrete form.

In a comparison of two concrete forms with identical cement/aggregrate/waste/water formulations, geometries, and volumes it would be expected that comparable pore spaces (i.e., having similar total volumes, surface area and pore size distributions) would develop as the concrete cured. However, if curing times are different, it would be expected that the matric potentials exhibited by the monoliths would be different. The net long-term effect of this initial difference in matric potential should, given sufficient contact with water, become insignificant for any or all of the following reasons:

1) wastes are "stored" following solidification and containerization prior to emplacement in the waste trench such that the concrete has an opportunity to cure "completely,"

2) wastes have been completely cured to an equilibrium pore water content, containerized and placed in a waste trench, and

3) wastes have been solidified, containerized and placed in a waste trench prior to "completely" curing but curing continues in the waste trench.

For the majority of concrete-solidified wastes in carbon steel outer containers, an induction period is expected to pass prior to breach of the outer container. Assuming the outer container was not breached on emplacement in the trench, initial breach would be accepted as a necessary occurrence for leaching to begin. During this time, some curing of the concrete could occur, given that the relative humidity of the trench environment were low enough. Once water infiltration of the solidified waste can occur, it will proceed to the exhaustion of either a) the available water, or b) the matric potential.

As can be seen in Figure 3.2.4, the volume of water influx can be calculated as the cross-sectional area of the corrosion pit (P_i) times the water flow (W_i) for the particular time, t_i. The incremental influx volumes are termed "ℓ_i". These accumulate in the waste package (in the waste itself) until, as discussed earlier, the void and pore space is filled. Until such a time, the model increments t_i and returns to calculate the next pit size and succeeding influx of water. As mentioned in the section on container corrosion, once a critical area of pitting has occurred (chosen here as equivalent to the drum top area) the value of P_i remains constant at whatever value the critical area is set to be.

Once the void and pore space volume of the waste is filled, the driving force (matric potential) for absorption of water is exhausted and the next drop of water that enters is seen as having to "push" an equivalent water volume (leachate) "out". It should be noted here that the assumption of pore and void space filling prior to exit of leachate is only one of a number of possibilities. Net water flow is driven by differences in potential which may take on a wide range of values. Saturation of pore spaces may, in many conceivable circumstances, not be necessary before leaching occurs.

An assumption implicit in the data generation presented in this section is that, between wetting stages (i.e., between t_i) no drying of the waste has occurred. In other words, all the water that has contacted the waste has stayed there as liquid water. A refinement to this would be to set a certain (technically justifiable) amount of water aside as subject to evaporation or drying from the waste. The net effect of this will be that it will take longer for the total accumulated ℓ_i to reach, and then exceed, the pore space volume (i.e., before removal or release of radionuclides could occur). This section is presenting the most conservative case (i.e., that leading to earliest release) while the opposite end of the range could also be accommodated. This would be the situation where the loss by evaporation or drying equalled the influx for the time interval chosen. In such a case, the soil and pore spaces of the waste would never completely fill.

For the set of circumstances where water can accumulate in the waste to the extent that it exceeds the void and pore space available, the next step represents removal of the radionuclide species residing at the sites accessible by leachant, i.e., at the surfaces of the void and pore spaces. As was mentioned in Section 3.2.2, the surface species modeled here are water-contact-soluble [described in F-(1) and F-(2)] and not subject to redeposition on or in the waste in the process of exiting the package [process F-(4)]. Diffusion processes, which are a necessary part of F-(3), may be modeled by a diffusion-system approach, while the redeposition (or inhibition of exit of leached species by plugging or other processes) may be modeled by a number of solubility-limited precipitation and/or adsorption isotherm approaches.

The actual surface species leaching model incorporates the concepts of a fixed "solution" volume (representing the solubilized surface species in the liquid volume prescribed by the void and pore spaces) and an incoming "rinse" volume (representing the influx of water for time increment, t_i). These are given the symbols V and ℓ_i, respectively. The total surface "source," $(X_o)_1$, is the total amount of radionuclide$_1$ present on the pore space surfaces, while $(X_o')_i$ is the net amount of radionuclide species remaining, i.e., after releases 1 to i have occurred. Each rinse depletes the "solution" volume by an amount expressed as:

$$R_i = \ell_i \frac{(X_o')_i}{V} \qquad (3.2.1)$$

where R_i is the amount of surface species radionuclides released in rinse i, for t_i,

ℓ_i is the "rinse" volume of water for t_i,

$(X_o')_i$ is the surface species source, and

V is the void and pore space "solution" volume.

It can be seen from this expression that the release represents the simplest case, independent of specific chemistry of the radionuclide, or of the leachant. Also, the replenishment of the "surface species" is not occurring.

For rinses of equal magnitude for successive t_i, the general formula for the incremental amount released has been calculated for a unit source, X_o, as [Kempf, 1983]:

$$IFR = z(1-z)^{n-1} \qquad (3.2.2)$$

where IFR is the incremental fraction released,

z is the ratio of the individual rinse volume to the total "solution" volume, and

n is the number of the rinse (equivalent to i).

This expression could be derived by virtue of the fact the successive rinses were equal in volume. For the present case, the rinse volumes continually increase with time up to a limit (discussed earlier) where the influx water volume becomes equal to a set, fixed area times the water flow (for a 55-gallon drum, this fixed drum top area is 2570 cm^2). After this point, the rinse volumes are equal to each other, given equal W_i. A similar general formula for this type of leaching model can not be derived when the ℓ_i are variable. An example of the expression used to calculate the amount released in the fourth rinse (after V has been accumulated) is:

$$R_4 = \frac{\ell_4 (X_o')_1}{V} - \frac{\ell_4 \ell_3 (X_o')_1}{V^2} + \frac{\ell_4 \ell_3 \ell_2 (X_o')_1}{V^3} - \frac{\ell_4 \ell_3 \ell_2 \ell_1 (X_o')_1}{V^4} \; . \qquad (3.2.3)$$

In calculating the incremental amounts released for this model, the individual ℓ_i have been calculated, then stored and accumulated. For each ℓ_i, an R_i has been calculated from:

$$R_i = \ell_i \frac{(X_o')_i}{V} \qquad (3.2.1)$$

where $(X_o')_i$ is an updated "source", i.e., for each release, R_i, the "source" has been appropriately decreased,

and other variables have been defined earlier.

The process of incremental releases continues until the test for depletion of the surface species "source" is positive:

$$\sum R_i \geq (X_o')_1 \qquad (3.2.4)$$

i.e., the cumulative release has become equivalent to the original amount present, $(X_o')_1$.

Once this has occurred, a subroutine for diffusion and solid dissolution can be called upon. For the data presented here, this total depletion of the original source, $(X_o')_1$, has been the stopping point.

As mentioned earlier in this section, several variables exist in this model:

k	= 0.095 or 0.95 ($cm/year^n$)
n	= 1 or 1/2
$X_o$total	= 1 (taken so that releases can be considered fractional releases)
$(X_o')_1$	= surface species "source" (see Section 3.2.2.2 for method of calculation) - dependent on void space fraction of the waste
W	= 1, 10, 100 (cm/year)

and

V	= 10%, 20%, 30%, 40% of a 55-gallon drum size.

3.2.4 Presentation of Data from the Model

Data have been generated for a series of sets of values assumed for the model variables. These sets have been organized into a matrix system as shown in Table 3.2.1. The values of k, n, and X_o determine the first digit of the data set while the second two digits are determined by W and V, respectively. For example, data set 312 corresponds to the following variable values:

k	= 0.95 ($cm/year^{1/2}$)
n	= 1/2
X_o	= 1
W	= 1 (cm/year)
V	= 42000 (cm^3)

This value of V is determined from an assumed 20% void space in a 210-liter (55-gallon) waste drum. All other values have been discussed previously.

These data sets have been labeled DSqmo where q, m, and o may take on the values 1, 2, or 3 according to Table 3.2.1. Plots of incremental and cumulative releases are given for DS1m1 and DS1m2. Figures 3.2.6 and 3.2.7 give the incremental and cumulative releases for DS1m1 (DS111, DS121, DS131) and Figures 3.2.8 and 3.2.9 give the incremental and cumulative releases for DS1m2 (DS112, DS122, DS132). These are for a variety of k, n, W and V combinations.

Figures 3.2.6 and 3.2.7 correspond to surface species leaching of a 55-gallon size waste with a 10% void space fraction. The values of k, n, X_o and V are held constant (see legends for the figures) and the variable is the water influx, W_i. The cummulative release reaches 0.15 because this is the corresponding surface species source for a 10% void waste.

The time interval to first release can be seen to vary with W, i.e., for W=100 the first release occurs about year 6, while for W=10 it is year 13 and for W=1, year 28. These times correspond to the amount of time needed to accumulate the void space volume of water (21000cm^3). Along with the variability of initial release times, it can be seen that the rate of incremental release is fastest for W=100 and slowest for W=1. This would be expected since a larger volume "rinse" should effectively deplete the solution faster than the smaller volume "rinse".

For the given conditions (k=0.95, n=1/2, etc.), W=100 leads to first release at year 6 and completion of surface species removal by year 10. For W=10 initial release starts at year 13 and continues until year 23. For W=1, initial release is at year 28 and continues until approximately year 55. A high water influx value leads to early, fast, relatively-high-valued release while a lower water influx takes longer to start release, the process is extended over a much longer period and the relative values of release are correspondingly lower.

It can be seen from Figures 3.2.8 and 3.2.9 that the relative shapes of incremental and cumulative releases (W=100,10,1) for 20% void space wastes are quite similar to those for 10% void space wastes. The total leached is correspondingly greater (0.25, see Section 3.2.2.2) and the first release times are later (W=100, 8 years; W=10, 16 years; W=1, 31 years). Also for 20% void fraction waste, the length of leaching is correspondingly longer (W=100, 5 vs. 4 years; W=10, 14 vs. 10 years; and W=1, 59 vs. 27 years).

It is clear from this cursory analysis of the results that a doubling of the void fraction does not result in a simple doubling of starting times or of total leaching times. This would indicate that the complexity of the conditions to be met in this surface species leaching model preclude the assignment of correlation factors at this point. An example can be made of the time to filling of the void space, V. If this interval is called t*, an expression for the filling time may be derived from:

Table 3.2.1 Parameter value sets for this model.

k = 0.95 n = 1 X_0 = 1	k = 0.095 n = 1 X_0 = 1	k = 0.95 n = 1/2 X_0 = 1	k = 0.095 n = 1/2 X_0 = 1
1	2	3	4

W	1	10	100
V	21000_{11}	21000_{21}	21000_{31} (10%)
V	42000_{12}	42000_{22}	42000_{32} (20%)
V	63000_{13}	63000_{23}	63000_{33} (30%)
V	84000_{14}	84000_{24}	84000_{34} (40%)

$$V(t) = \int_0^{t^*} W\, A(t')\, dt'$$

$$= \int_0^{t^*} W \cdot \pi k^2 (t')^{2n}\, dt'$$

$$= W\pi k^2 \int_0^{t^*} (t')^{2n}\, dt'$$

$$= W\pi k^2 \frac{t^{2n+1}}{2n+1} \Bigg]_0^{t^*}$$

$$\text{so, } t^* = \left[\frac{V(2n+1)}{W\pi k^2} \right]^{1/(2n+1)}$$

This expression explicitly shows the nonlinearity between time to first release and the other parameters.

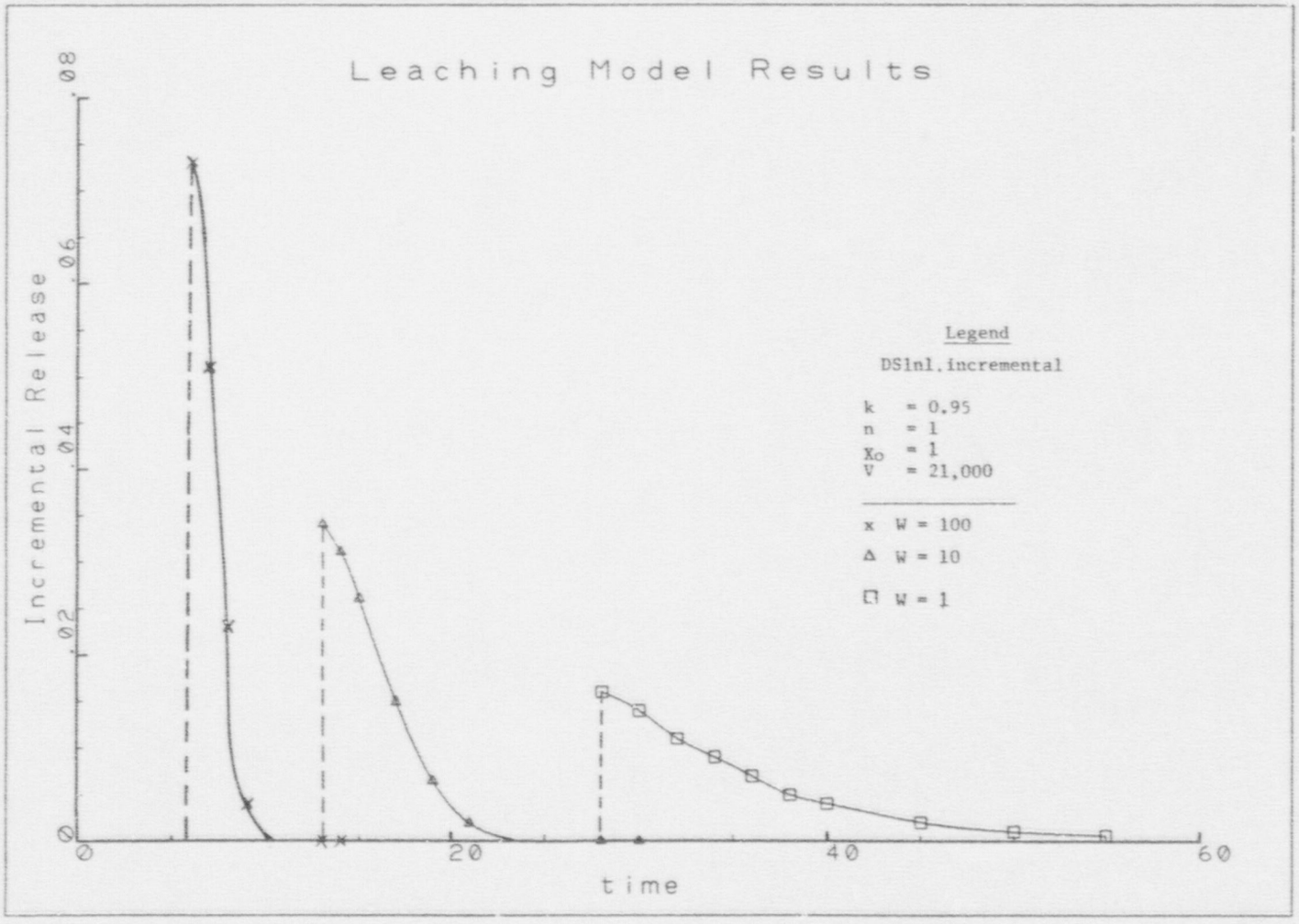


Figure 3.2.6 Leaching model results for data set inl incremental.

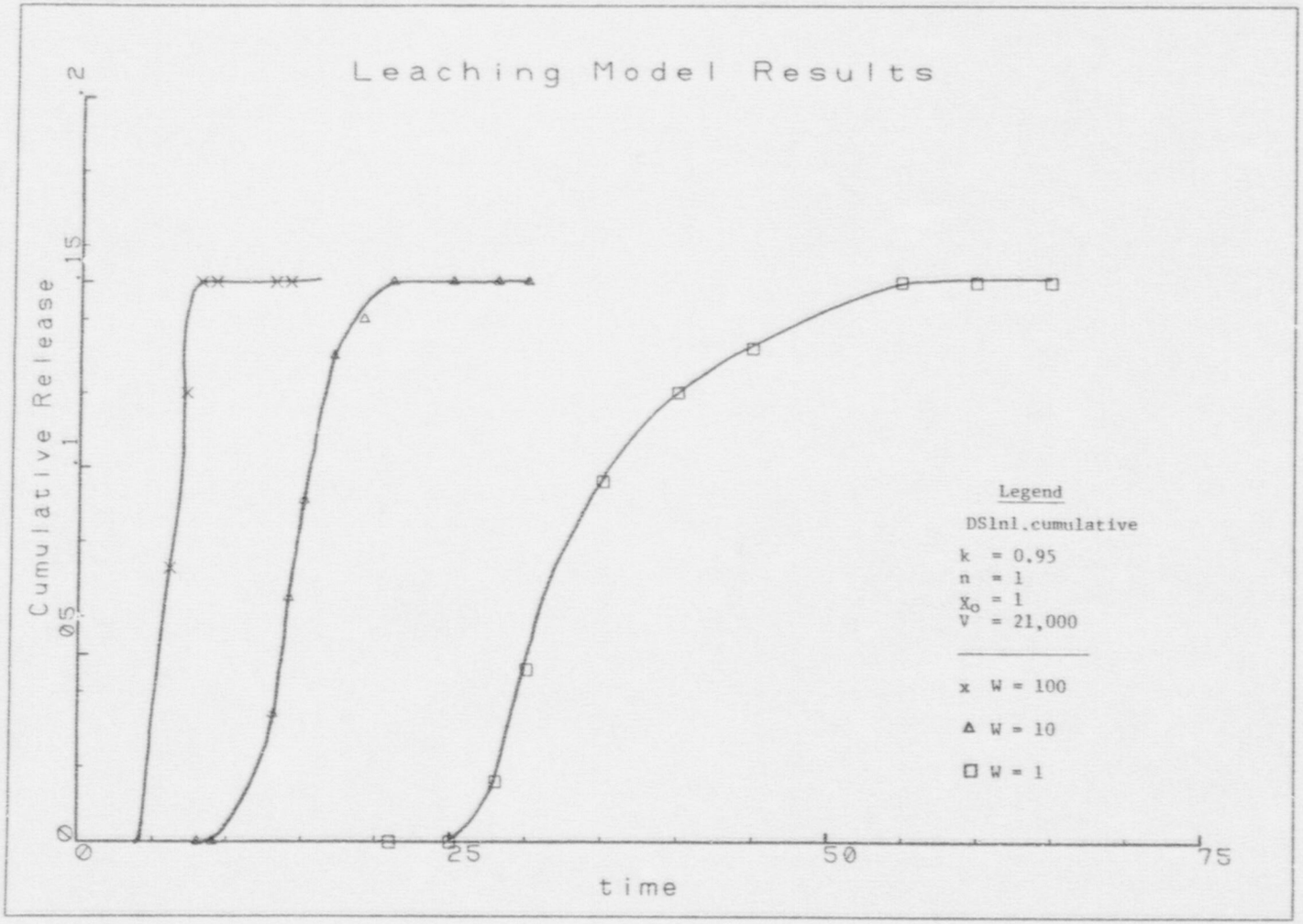


Figure 3.2.7 Leaching model results for data sets lnl cumulative.

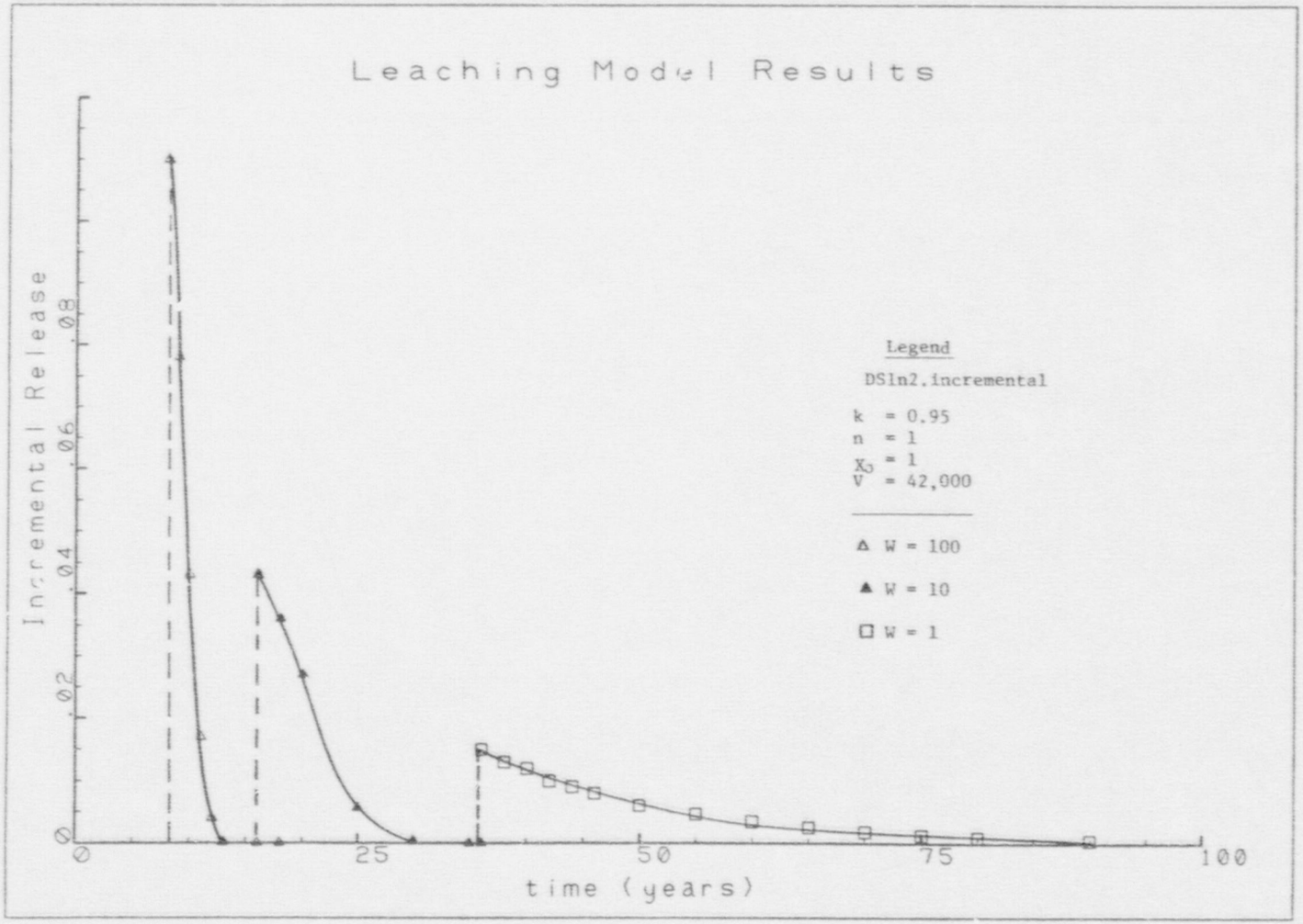


Figure 3.2.8 Leaching model results for data sets ln2 incremental.

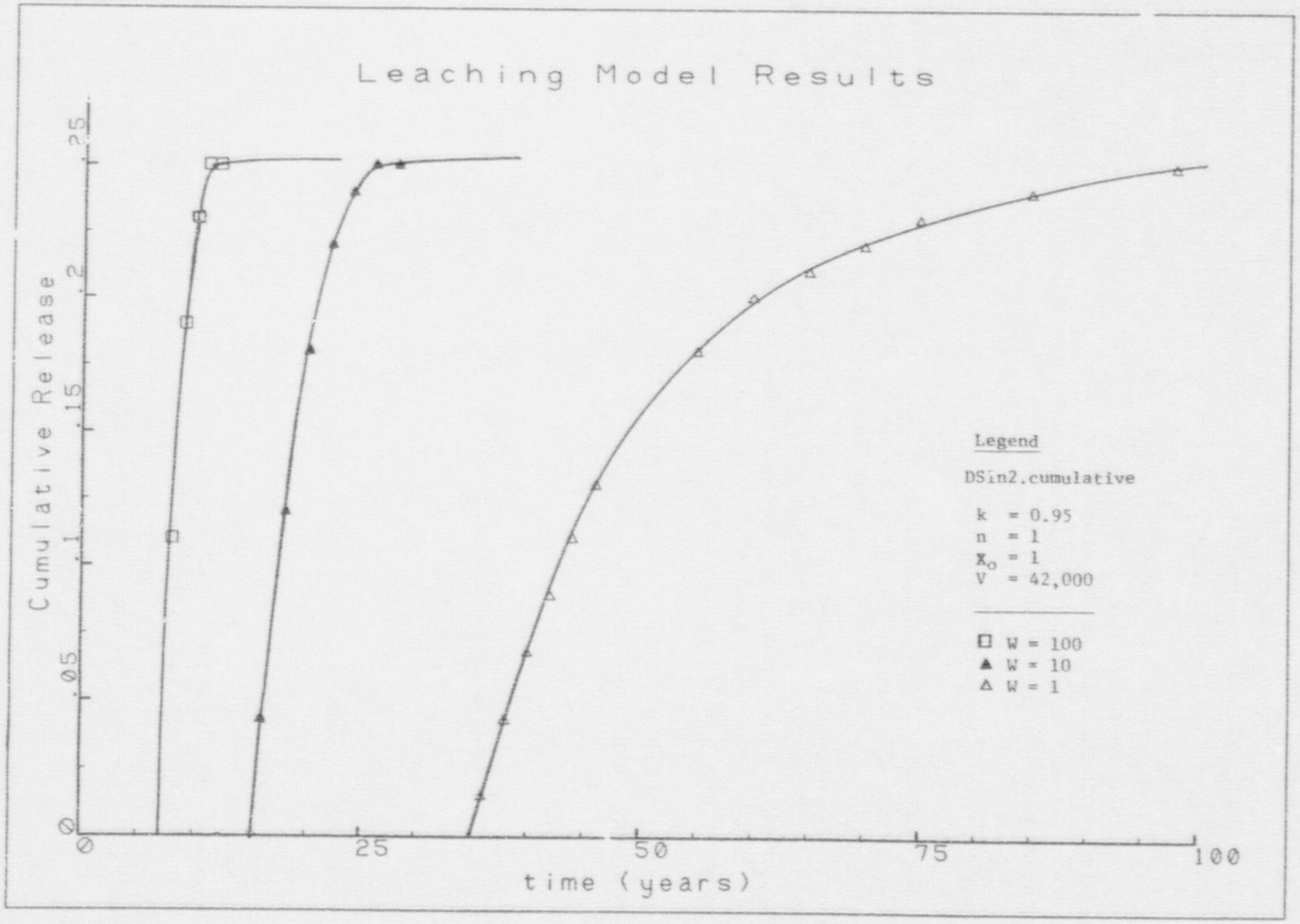


Figure 3.2.9 Leaching model results for data sets ln2 cumulative.

3.3 Modeling Contaminant Transport Through the Disposal Trench

The ultimate concern for disposal of radioactive waste by shallow-land burial is the potential for the waste radionuclides to contact the public. In this source term project, the first stage in this process, release from the disposal trench is examined. For most radionuclides of concern, migration with the water will be the principal path by which they exit the trench.

This section presents the basic approach used in modeling radionuclide transport in an unsaturated porous medium. The remainder of this section presents a discussion of the basic transport equation along with the associated initial and boundary conditions, and solution techniques for the contaminant transport equation.

3.3.1 Radionuclide Transport Equation

As with the water flow equations, the starting point is the mass balance equation for each radionuclide. In deriving this equation, it is assumed that the air phase does not contribute to the movement of the nuclides. This is reasonable except for those radionuclides which can exist in the gas phase, such as tritium and carbon. Further, it is assumed that the soil and waste form are immobile. Thus, radionuclides in these media are fixed spatially. The soil and waste form act as sources and sinks of nuclides through their interaction with the ground water. Specifically, the waste form acts as a source due to the leaching process and the soil can act as a source or sink due to chemical interactions with the ground water (e.g. sorption). With these assumptions, the mass balance equation for each radionuclide takes the form [Oster, 1982]:

$$\frac{\partial}{\partial t}\left[(1-\phi)\ \rho_m\ C_m\right] + \frac{\partial}{\partial t}\left[\theta\ \rho_w\ C_w\right] = \nabla\cdot\left[\theta\ \rho_w\ D_w\ \nabla C_w\right] - \nabla\cdot\left[\rho_w\ C_w\ V_w\right] - S_c \qquad (3.3.1)$$

Where: ϕ = porosity,
ρ = density of the phase (ML^{-3})
C = concentration of the phase (ML^{-3})
θ = volumetric moisture content (L^0)
D = dispersion coefficient (L^2T^{-1}),
V = pore water velocity (LT^{-1}),
S = source or sink of radionuclides
M = mass
L = length, and
T = time

the subscripts w and m refer to the water and matrix (soil) phases.

A common approach used to relate the concentration on the soil to the concentration in the solution is to assume that they are related by an isotherm of the form:

$$C_m = k_d \rho_w C_w \tag{3.3.2}$$

Here, k_d is the distribution coefficient which is defined as:

$$k_d = \frac{\text{mass of species adsorbed on the solid phase per unit mass of solid}}{\text{concentration of species in solution}}$$

Using this expression and assuming the water is incompressible, Equation (3.3.1) becomes:

$$\frac{\partial}{\partial t} (\theta R C_w) = \nabla \cdot \theta D_w \nabla C_w - \nabla \cdot V C_w - S_c \tag{3.3.3}$$

Where the retardation factor R is defined as:

$$R = 1 + \rho_b k_d / \theta \tag{3.3.4}$$

Here, ρ_b is the bulk density of the soil and equals $(1-\theta)\rho_m$.

Use of a retardation factor to represent all of the chemical interactions between the soil and the water is a simplistic approach that is widely used because it linearizes the mass transport equation and makes its solution numerically easier. However, this model is only valid if the solution contains only trace amounts of the contaminant and competition for sorption sites between different contaminants is negligible. Several more elaborate models have been developed (for example, Carnahan, 1984) which include sorption, ion exchange, dissolution and precipitation phenomena. If it is felt that the use of a retardation factor is inadequate, the more general equation, (3.3.1) will be used with appropriate models for the soil water interaction term.

The source/sink term in this equation will include a model for release from the waste form (Section 3.2). The level of sophistication used in the leach model could range from simple empirical expressions, (e.g., specifying the leach rate as linearly proportional to time or to the square root of time), to more detailed mechanistic descriptions of leaching.

The initial condition required for solution of Equation (3.3.3) is the concentration in the water of each radionuclide at each spatial location within the trench. For this problem, the system should be free of each radionuclide. Contaminants will enter the system from the leaching of the waste forms.

The boundary conditions for the contaminants take the same mathematical form as for the hydraulic potential in the water flow equation, Equation (3.1.7). That is, either the concentration:

$$C(0,t) = C_b(t) \tag{3.3.5}$$

or the flux

$$\left(-\theta D\frac{dC}{\partial x} + VC\right)\Big|_{x=0} = q_b(t)\ C_b(t) \quad q_b>0 \tag{3.3.6}$$

$$= 0 \quad q_b<0$$

may be specified as a function of time. Where $q_b(t)$ is the volumetric flux of water and C_b is the concentration of the radionuclide in the infiltrating water at the boundary under consideration.

If the concentration at each boundary is specified to be zero, this should produce the highest flux of contaminants out of the system because the concentration gradients will be at their maximum value. However, recalling that an objective of this project is to provide both the flux and the concentration of contaminants leaving the system, the choice of zero concentration as a boundary condition may not be appropriate.

3.3.2 Solution Techniques for the Radionuclide Transport Equation

The mathematical form of the radionuclide transport equation is similar to that of the water flow equation. Both are elliptic, partial differential equations. Therefore, similar solution techniques are used to solve both equations. That is, the finite difference and finite element methods are the most commonly used solution techniques.

The major differences between the two equations are the non-linearity of the water flow equation due to the material properties depending on moisture content, and the advection term in the radionuclide transport equation.

In general, the radionuclide transport equation may be non-linear due to concentration-dependent material properties, such as dispersivity or soil-water interactions. However, in the case of Equation (3.3.3), the assumption of a linear sorption isotherm (Equation 3.3.2) and constant material properties removes the non-linearities from the equation.

The advection term does not add any conceptual difficulties to the numerical solution of the radionuclide transport equation. However, there is the practical difficulty that numerical schemes that work well for dispersion-dominated flow do not necessarily work well for advection-dominated flow. This problem can be overcome by an automated procedure that selects the proper scheme based on the relative importance of these two processes.

As with the water flow equations, there are many computer codes which have been written to predict solute transport. A compilation of these codes can be found in van der Heijde [van der Heijde, 1985]. Many of these codes are documented and available to the public. The possibility of obtaining an existing code and modifying the code to fit the needs of the source term project is being explored.

4.0 CONCLUSIONS AND FUTURE WORK

The objective of this project is to develop a methodology to predict the release rate of radionuclides from a shallow land disposal unit. Work to date has focused on reviewing disposal practices, modeling, and experimental work relevant to the project. Further, the framework for modeling of radionuclide release and some preliminary models have been developed. Based on this work, the following conclusions have been drawn and areas for future work have been identified.

- The placement of low-level waste shallow land burial sites above the water table ensures that, for the vast majority of the time, the wastes will be in unsaturated conditions. Modeling of water flow in porous media to the present time has been dominated by an assumption of saturated conditions. Modeling for unsaturated conditions is, in general, and specifically for water flow, a more difficult and relatively new undertaking.

- It is assumed that two general types of low-level waste packages are being generated subsequent to the issuance of 10 CFR Part 61; radionuclide release rates from these wastes are being modeled in the source term project. These two main waste types are unstabilized Class A and stabilized Class B and C. Each of these broad categories may be broken down into a number of subgroups. Initially at least, the source term modeling effort will concentrate on wastes that may be categorized as porous, contained in corrodible outer containers. Then, wastes solidified in encapsulating agents (bitumen), and wastes placed in high integrity containers will be considered.

- Little or no specific information is available on the particular chemical forms of waste radionuclides. This is not surprising since generators are generally more concerned about qualitative and quantitative characterization of products as opposed to wastes. However, radionuclide releases are being modeled in this project and release mechanisms are quite dependent on chemistry. It is also known that prediction of speciation of these released radionuclides is dependent on knowledge of the speciation in the waste itself (as well as subsequent to solidification or otherwise effected through stabilization). At this point, modeling is expected to proceed to the point of gross radionuclide species identification, i.e., prediction of hydrated anionic or cationic states, mainly as a function of ranges in pH.

Future Work

Models are needed for water flow, container degradation, waste form leaching, and radionuclide transport. Whenever possible, existing models will be adapted to the needs of the source term project. Several computer codes that predict water flow and/or radionuclide transport in an unsaturated porous medium exist. It is likely that both of these processes will be modeled by existing codes modified for this project. Container degradation and waste form leaching models tend to be empirical and are often not applicable to wastes in shallow land disposal. Models for these two processes will be developed as part of the source term project.

Knowing the water flow rate is essential in determining waste form leaching and radionuclide transport. Therefore, the most immediate need of the source term project is to obtain a method for estimating water flow.

Modeling water flow at this point must involve assumptions that homogenize the system. Specifically, the backfilling around waste containers is assumed to be reasonably uniform, and due to the long time period, precipitation events are averaged over time. Future work might include determination of the influence of these assumptions on water flow through modeling of effects of voids and fractures in the disposal unit and varying the time over which precipitation events are averaged.

Trench inventories need to be established by waste types, volume, classes, etc. to allow some type of comparison to the assumed inventories/ waste package categories. This would assure that the modeling cases cover at least a majority of the waste package possibilities (and therefore, might give a reasonable estimate of radionuclide release quantities).

Details of radionuclides' chemical behavior in solidification agents and also that occurring in the process of permeation through polymeric materials as well as passing through soils need to be examined.

5.0 REFERENCES

[Abeele, 1985]
Abeele, W. V., "Consolidation and Shear Failure Leading to Subsidence and Settlement, Part 1," Los Alamos National Laboratory, LA-10261-MS, 1985.

[ANS, 1984]
American Nuclear Society, "Measurement of the Leachability of Solidified Low-Level Radioactive Wastes," American Nuclear Society Standards Committee, Final Draft of Standard ANS-16.1 (1984).

[Arora, 1985]
Arora, H., and Dayal, R., "Properties of Radioactive Wastes and Waste Containers, Progress Report July - September, 1985," Brookhaven National Laboratory, December, 1985.

[Arora, 1986]
Arora, H., and Dayal, R., "Leaching Studies of Cement-Based Low-Level Radioactive Waste Forms," Brookhaven National Laboratory, NUREG/CR-4756, BNL/NUREG-52026, October, 1986.

[Barletta, 1983]
Barletta, R. E., et. al., "Solidification of Irradiated Epicor-II Waste Products," NUREG/CR-2969, May, 1983.

[Bennett, 1984]
Bennett, R. D. et. al., "Alternative Methods for the Disposal of Low-Level Radioactive Wastes Task 1: Description of Methods and Assessment of Criteria," Vol. 1, Vicksburg, Mississippi: U.S. Army Engineer Waterways Experiment Station, NUREG/CR-3774, 1984.

[Blanco, 1966]
Blanco, R. E., et. al., "Recent Developments in Treating Low- and Intermediate-Level Waste in the USA," Practices in the Treatment of Low- and Intermediate-Level Radioactive Wastes, IAEA and ENEA-(SM-71/26), p. 793-839, 1966.

[Campana, 1982]
Campana, R. J., et. al., "Corrosion and Reliability Monitoring," Report GA-C16932 on GA Technologies Project No. 3386, November, 1982.

[Carnahan, 1984]
Carnahan, C.L., et. al., "Verification and Improvement of Predictive Algorithms for Radionuclide Migration," LBL-17133, UC-70, 1984.

[Colombo, 1979]
Colombo, P., and Neilson, R. M. Jr., "Properties of Radioactive Wastes and Waste Containers, First Topical Report," NUREG/CR-0619, BNL-NUREG-50957, August 1979.

[Dayal, 1983]
Dayal, R., Arora, H., and Morcos, N., "Estimation of Cesium-137 Release From Waste/Cement Composites Using Data From Small-Scale Specimens, Topical Report," NUREG/CR-3382, BNL-NUREG-51690, July 1983.

[Dayal, 1985a]
Dayal, R., and Arora, H., "Properties of Radioactive Wastes and Waste Containers, Progress Report, October - December, 1984," Brookhaven National Laboratory, April, 1985.

[Dayal, 1985b]
Dayal, R., and Arora, H., "Properties of Radioactive Wastes and Waste Containers, Progress Report, April - June, 1985," Brookhaven National Laboratory, August, 1985.

[Dormuth, 1981]
Dormuth, K. W., and Sherman, G. R., "SYVAC - A Computer Program for Assessment of Nuclear Fuel Waste Management Systems, Incorporating Parameter Variablity," Research Report AECL 6814, August, 1981.

[Dougherty, 1985a]
Dougherty, D. R., and Colombo, P., "Leaching Mechanisms of Solidified Low-Level Waste: The Literature Survey," Brookhaven National Laboratory, BNL 51899, June, 1985.

[Dougherty, 1985b]
Dougherty, D. R., Fuhrmann, M. and Colombo, P., "Accelerated Leach Test(s) Program Annual Report," Brookhaven National Laboratory, BNL 51955, September, 1985.

[Eicholz, 1985]
Eicholz, G. C., et. al., "Waste Migration Under Unsaturated Flow Conditions," Waste Management-85, eds. R. G. Post and M. E. Wacks, p. 215, 1985.

[Electrowatt, 1983]
Electrowatt Engineering Services (UK) Ltd., "Development of Vault Model "VERMIN" for Post Closure Behaviour of Repositories for Non-Heat Generating Radioactive Wastes," Department of Environment, DOE/RW/84.034, EWE-FWM-06-2518, 1983.

[Gause, 1983a]
Gause, E.P., et. al., "Characterization of the Radioactive Large Quantity Waste Package of the Union Carbide Corporation," Brookhaven National Laboratory, NUREG/CR-2870, BNL-NUREG-51576, 1983.

[Gause, 1983b]
Gause, E.P., et. al., "Characterization of the Class B Stable Radioactive Waste Packages of the New England Nuclear Corporation," Brookhaven National Laboratory, NUREG/CR-3018, BNL-NUREG-51607, 1983.

[General Research Corporation, 1980]
General Research Corporation, "Study of Chemical Toxicity of Low-Level Wastes," Vol 1., NUREG/CR-1793, 1980.

[Haggblom, 1979]
Haggblom, H., and Kjelbert, N., "Calculations of Radioactive Nuclide Migration From a Repository for Reactor Wastes," Prav. 1.27, Studsvik Energiteknik AB, Nykoping, Sweden, 1979.

[Hespe, 1971]
Hespe, E. D., "Leach Testing of Immobilized Radioactive Waste Solids. A Proposal for a Standard Method," Atomic Energy Review, 195-207, 1971.

[Hillel, 1971]
Hillel, D., Soil and Water, Academic Press Inc., New York, 1971.

[Hillel, 1983]
Hillel, D., Introduction to Soil Physics, New York, Academic Press, 1983.

[Hung, 1983]
Hung, C. Y., Meyer, G. L., and Rogers, V. C., "Use of PRESTO-EPA Model in Assessing Health Effects from Land Disposal of LLW to Support EPA's Environmental Standards," CONF-8308106, Fifth Annual DOE Low-Level Waste Management Participants Meeting, p. 529-541, 1983.

[IGWMC, 1986]
International Ground Water Modeling Center, "Price List of Publications and Services Available from IGWMC," Holcomb Research Institute, Butler University, Indianapolis, Indiana, September, 1986.

[Intera, 1983]
INTERA Environmental Consultants, Inc., "WAPPA: A Waste Package Performance Assessment Code," ONWI-452, April, 1983.

[Jones, 1978]
Jones, T. L., "Sediment Moisture Relations: Lysimeter Project 1976-1977 Water Year," Rockwell International, Rockwell Hanford Operations, RHO-ST-15, June, 1978.

[Kempf, 1983]
Kempf, C. R., Memo to File, Brookhaven National Laboratory, "Model for Releases of Cs-137 and Sr-90 from Wastes Shipped by Minnesota Mining and Manufacturing (3M) - Buried at Sheffield Site, 1967-1971, Trenches 1 and 2," December 29, 1983.

[Kempf, 1984a]
Kempf, C. R., et. al., "Characterization of the Low-Level Radioactive Wastes and Waste Packages of General Electric Vallecitos Nuclear Center," NUREG/CR-3864, BNL-NUREG-51791, 1984.

[Kempf, 1984b]
Kempf, C. R., "Characterization of the Low-Level Radioactive Wastes and Waste Packages of the Minnesota Mining and Manufacturing Company," Brookhaven National Laboratory, Draft Report, BNL-NUREG-34392, March 1984.

[Kempf, 1986]
Kempf, C. R., Memo to File, Brookhaven National Laboratory, "Development Considerations for a Model to be Used for Leaching of a Porous Waste Form," June 27, 1986.

[Kennedy, 1986]
Kennedy, W. E., "Intruder Dose Pathway Analysis for the Onsite Disposal of Radioactive Wastes: The Onsite/Maxi-1 Computer Program," NUREG/CR-3620, Suppl. 1, 1986.

[Kincaid, 1984]
Kincaid, C. T., Morrey, J. R., and Rogers, J. E., "Geohydrochemical Models for Solute Migration. Volume 1: Process Description and Computer Code Selection," EPRI EA-3417, February, 1984.

[King, 1985]
King, C. M., et. al., "The Savannah River Laboratory DOSTOMAN Code - A Compartmental Pathways Computer Model of Contaminant Transport," CONF-8509121, Seventh Annual DOE Low-Level Waste Management Participants Meeting, p. 814-828, 1986.

[Lester, 1981]
Lester, D., et. al., "System Analysis of Shallow Land Burial," Science Applications, Inc., NUREG/CR-1963, Vol. 3, 1981.

[Liakopoulos, 1965]
Liakopoulos, A.C., "Transient Flow Through Unsaturated Porous Media," Ph.D. Dissertation, Department of Civil Engineering, University of California, Berkeley, 1965.

[MacKenzie, 1985]
MacKenzie, D. R., et. al, "Evaluation of the Radioactive Inventory In, and Estimation of Isotopic Release From the Waste in Eight Trenches at the Sheffield Low-Level Waste Burial Site," NUREG/CR-3865, BNL-NUREG-51792, January, 1985.

[MacKenzie, 1986]
MacKenzie, D. R. , et. al., "Preliminary Assessment of the Performance of Concrete as a Structural Material for Alternative Low-Level Radioactive Waste Disposal Technologies," NUREG/CR-4714, BNL-NUREG-52016, 1986.

[Matsuzuru, 1979]
Matsuzuru, H., Moriyama, N., and Ito, A., "Leaching Behavior of Tritium From a Hardened Cement Paste," Annals of Nuclear Energy, V. 6, p. 417-423, 1979.

[MCC, 1981]
Materials Characterization Center, "Nuclear Waste Material Handbook," DOE/TIC-11400, 1981.

[McIntyre, 1986]
McIntyre, P. F., "Performance of Special Waste Form Lysimiters and Waste Migration at a Humid Site," Presented at the Eighth Annual Participants' Information Meeting DOE Low-Level Waste Management Program, Conf-860990, Denver, CO, September, 1986.

[McNeil, 1986]
McNeil, M. B., "Statistical Issues in Pitting Corrosion of Carbon Steel," Presented at the Materials Reasearch Society Annual Meeting in Boston, MA, December, 1986.

[Mendel, 1984]
Mendel, J. E., editor, "Final Report of the Defense High-Level Waste Leaching Mechanisms Program," PNL-5157, August, 1984.

[Mezga, 1984]
Mezga, L. J., "The Role of Trench Caps in the Shallow Land Burial of Low-Level Wastes," ORNL/TM-9156, 1984.

[Moore, 1977]
Moore, J. G., Godbee, H. W., and Kibbey, A. H., "Leach Behavior of Hydrofracture Grout Incorporating Radioactive Wastes," Nuclear Technology, Vol. 32, p. 39-52, Jan. 1977.

[Neumann, 1974]
Neumann, S. P., Feddes, R. A., and Bresler, E., "Finite Element Simulation of Flow in Saturated-Unsaturated Soils Considering Water Uptake by Plants," Third Annual Report, Project ALO-SWC-77, Hydrodynamics and Hydraulic Engineering Laboratory Technion, Haifa, Israel, 1974.

[Neville, 1981]
Neville, A. M., Properties of Concrete, Third Edition, p. 32, Pitman Publishing, Limited, London, 1981.

[Oblath, 1985]
Oblath, S. B., and Grant, M. W., "Special Wasteform Lysimeters Initial Three-Year Monitoring Report," Savannah River Laboratory, DP-1712, 1985.

[Ohnuki, 1986]
Ohnuki, T., "Migration of Radioactive Cobalt Dissolved in the Solution of pH 12 Through Sandy Soil Layer," Nuclear and Chemical Waste Management, Vol. 6, p. 175-180, 1986.

[Oster, 1982]
Oster, C. A., "Review of Groundwater Flow and Transport Models in the Unsaturated Zone," NUREG/CR-2917, PNL-4427, 1982.

[Otis, 1986]
Otis, M. D., "Safety Assessment of Alternatives to Shallow Land Burial of Low Level Radioactive Waste: Volume 1. Failure Analysis of Engineered Barriers," NUREG/CR-4701, EGG-2465 (1986).

[Pescatore, 1983]
Pescatore, C., "Mechanistic Modeling of Nuclear Waste Form Leaching by Aqueous Solutions," Ph.D. Dissertation, Dept. of Nuclear Engineering, University of Illinois, 1983.

[Phillips, 1984]
Phillips, D. C., et. al., "The Effects of Radiation on Organic Matrix Waste Forms in Testing," Evaluation and Shallow Land Burial of Low and Medium Level Radioactive Waste Forms, W. Krischer and R. Simon, editors, Harwood Academic Publishers, p. 183-199, 1984.

[Pittiglio, 1986]
Pittiglio, C. L., and Tokar, M., "NRC Perspective on Alternative Disposal Methods," Presented at the Eighth Annual Participants' Information Meeting DOE Low-Level Waste Management Program, Conf-860990, Denver, CO, September, 1986.

[Polzer, 1986]
Polzer, W. L., et. al., "Modeling Study of Solute Transport in the Unsaturated Zone: Information and Data Sets," NUREG/CR-4615, LA-10730-MS, Vol. 1, 1986.

[Romanoff, 1957]
Romanoff, M., Underground Corrosion, NBS Circular 579 PB-168350, April, 1957.

[Saltelli, 1984]
Saltelli, A., Avogadro, A., and Bidoglio, G., "Americium Filtration in Glauconitic Sand Columns," Nuclear Technology, Vol. 67, p. 245-254, November, 1984.

[Sastre, 1986]
Sastre, C., Pescatore, C., and Sullivan, T., "Waste Package Reliability," Brookhaven National Laboratory, NUREG/CR-4509, BNL-NUREG-51953, February, 1986.

[Schulz, 1983]
Schulz, R. K., "Study of Unsaturated Zone Hydrology at Maxey Flats," CONF-8308106, Fifth Annual DOE Low-Level Waste Management Participants Meeting, p. 674-692, 1983.

[Schulz, 1986]
Schulz, R. K., and Ridky, R. W., "Control of Water Infiltration Through SLB Trench Covers," CONF-8509121, Seventh Annual DOE Low-Level Waste Management Participants Meeting, p. 122-139, 1986.

[Skaggs, 1986]
Skaggs, R. L., and Walter, M. B., "Summary: Special Waste Form Lysimeters - Arid Program," presented at the Eighth Annual DOE Low-Level Waste Management Participants Meeting, Denver, CO, 1986.

[Skryness, 1982]
Skryness, R., "Overview of Design Considerations for Effective Trench Covers." Proc. of the Symposium on Low-Level Waste Disposal NUREG/CP-0028, Vol. 3, p. 277, (1982).

[Soo, 1986]
Soo, P. , et. al, "The Effects of Environment and Gamma Irradiation on the Mechanical Properties of High Density Polyethylene," NUREG/CR-4607, BNL-NUREG-51991, 1986.

[Stephens, 1986]
Stephens, K., et. al., "Methodologies for Assessing Long-Term Performance of High-Level Radioactive Waste Packages," Aerospace Corporation, NUREG/CR-4477, ATR-85 (5810-01) 1ND, 1986.

[Stone, 1986]
Stone, J. A., et. al., "Waste Migration Studies at the Savannah River Plant Burial Ground," CONF-8509121, Seventh Annual DOE Low-Level Waste Management Participants Meeting, p. 110-121, 1986.

[Sullivan, 1985]
Sullivan, T., et. al., "Low-Level Waste Source Term Evaluation: Progress Report, June - September, 1985," Brookhaven National Laboratory, November, 1985.

[Sullivan, 1986]
Sullivan, T., et. al., "Low-Level Waste Source Term Evaluation: Progress Report, April - June, 1986," Brookhaven National Laboratory, August, 1986.

[van der Heijde, 1985]
van der Heijde, P. K. M., et. al., "Groundwater Management: The Use of Numerical Models." Water Resourc. Monogr. 5, 2nd edition. Washington, D. C. Am. Geoph. Union, 1985.

[Walton, 1986]
Walton, J. C., and Sagar, B., "A Corrosion Model for Nuclear Waste Containers," Presented at the Materials Research Society Meeting in Boston, MA, December, 1986.

[Wilhite, 1986]
Wilhite, E. L., "Waste Salt Disposal at the Savannah River Plant," Savannah River Laboratory, DP-MS-85-164, presented at the ANS International Meeting: Low-, Intermediate- and High-Level Waste Management and Decontamination and Decommissioning, Niagara Falls, NY, 1986.

[Yeh, 1980]
Yeh, G. T., and Ward, D. S., "FEMWATER: A Finite-Element Model of Water Flow Through Saturated-Unsaturated Porous Media," Oak Ridge National Laboratory, ORNL-5567, 1980.

[Yeh, 1981]
Yeh, G. T., and Ward, D. S., "FEMWASTE: A Finite-Element Model of Waste Transport Through Saturated-Unsaturated Porous Media," Oak Ridge National Laboratory, ORNL-5601, 1981.

DISTRIBUTION

Dr. Marcel P. Bergeron
Battelle - Pacific Northwest Laboratory
P. O. Box 999
Richland, WA 99352

Dr. David Brooks
U. S. Nuclear Regulatory Commission
Nuclear Materials Safety & Safeguards
Mail Stop 623 SS
Washington, DC 20555

Dr. Frank A. Costanzi
U. S. Nuclear Regulatory Commission
Office of Nuclear Regulatory Research
Waste Management Branch
Div. of Radiation Programs & Earth Sciences
Mail Stop 007 NL
Washington, DC 20555

Mr. T. M. Gilliam
Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, TN 37831

Dr. H. W. Godbee
Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, TN 37831

Dr. C. Hung
Low-Level Waste Standards
Office of Radiation Programs (ANR-46)
U. S. Environmental Protection Agency
Washington, DC 20460

Mr. T. Johnson
U. S. Nuclear Regulatory Commission
Waste Management/Engineering Branch
Div. of Waste Management
Office of Nuclear Materials Safety &
Safeguards
Mail Stop 623 SS
Wasnington, DC 20555

Dr. Linda A. Kovach
U. S. Nuclear Regulatory Commission
Office of Nuclear Regulatory Research
Waste Management Branch
Mail Stop 5650 NL
Washington, DC 20555

Dr. Patrick F. McIntyre
E. I. Du Pont de Nemours & Company
Savannah River Plant
Bldg. 773-43A
Aiken, SC 29808

Dr. Michael B. McNeil
U. S. Nuclear Regulatory Commission
Office of Nuclear Regulatory Research
Waste Management Branch
Division of Radiation Programs &
Earth Sciences
Mail Stop 1130 SS
Washington, DC 20555

Dr. Tin Mo
U. S. Nuclear Regulatory Commision
Office of Nuclear Materials & Safeguards
Division of Waste Management
Waste Management/Engineering Branch
Mail Stop 623-SS
Washington, DC 20555

Dr. Leroy S. Person
U. S. Nuclear Regulatory Commission
Office of Nuclear Material Safety & Safeguards
Division of Waste Management
Low-Level Waste & Uranium Recovery Projects
Branch
Mail Stop 623 SS
Washington, DC 20555

Dr. Jacob Philip
U. S. Nuclear Regulatory Commission
Division of Engineering Safety
Waste Management Technology Section
Rm. 263
Washington, DC 20555

Mr. R. J. Serne
Pacific Northwest Laboratories
P. O. Box 999
Richland, WA 99352

Dr. M. Tokar
U. S. Nuclear Regulatory Commission
Office of Nuclear Materials Safety & Safeguards
Division of Waste Management
Mail Stop 623 SS
Washington, DC 20555

Dr. Marcia B. Walter
Battelle - Pacific Northwest Laboratory
P. O. Box 999
Richland, WA 99352

Dr. E. L. Wilhite
E. I. Du Pont de Nemours & Co.
Savannah River Laboratory
Bldg. 773-43A
Aiken, SC 29808

BNL Distribution

Peter Colombo - 703
Dave Dougherty - 703
All Nuclear Waste Management Division Staff Members

NRC FORM 335 (2-84) NRCM 1102, 3201, 3202

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

SEE INSTRUCTIONS ~~ON THE~~ REVERSE

1. REPORT NUMBER (Assigned by TIDC, add Vol. No., if any)
NUREG/CR-4897
BNL-NUREG-52066

2. TITLE AND SUBTITLE
Low-Level Waste Source Term Evaluation: Review of Published Modeling and Experimental Work, and Presentation of Low-Level Waste Source Term Modeling Framework and Preliminary Model Development

3. LEAVE BLANK

4. DATE REPORT COMPLETED
MONTH: February YEAR: 1987

5. AUTHOR(S)
Terrence Sullivan and C. Ruth Kempf

6. DATE REPORT ISSUED
MONTH: April YEAR: 1987

7. PERFORMING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
Brookhaven National Laboratory
Upton, New York 11973

8. PROJECT/TASK/WORK UNIT NUMBER

9. FIN OR GRANT NUMBER
A-3276

10. SPONSORING ORGANIZATION NAME AND MAILING ADDRESS (Include Zip Code)
Division of Waste Management
Office of Nuclear Regulatory Research
U. S. Nuclear Regulatory Commission
Washington, DC 20555

11a. TYPE OF REPORT
Formal

b. PERIOD COVERED (Inclusive dates)
6/85 - 12/86

12. SUPPLEMENTARY NOTES

13. ABSTRACT (200 words or less)

ABSTRACT

This report contains information on the efforts performed to date on the Low-Level Waste Source Term Evaluation Project, the objective of which is development of a model to predict radionuclide release rates from a low-level waste disposal unit. The approach for model development has been based on a compartmentalized scheme focused on the four major processes of water flow, container degradation, waste leaching and waste radionuclide transport to the trench boundaries. This stage of the project is focused primarily on modeling release rates from shallow land burial as currently practiced. Research efforts to this point include characterization work (of burial trenches themselves, of soils and structural features, and of waste forms and containers), review of published modeling work, review of several waste package performance system models, and development of original container degradation and waste leaching models. Characterization of the wastes, containers, and of the site (trench soils and structure) has been based on the premise that NRC guidance has been put into effect.

14. DOCUMENT ANALYSIS - a. KEYWORDS/DESCRIPTORS
Low-level waste, source term, modeling, release rates, disposal trench

b. IDENTIFIERS/OPEN-ENDED TERMS

15. AVAILABILITY STATEMENT
unlimited

16. SECURITY CLASSIFICATION
(This page) unclassified
(This report) unclassified

17. NUMBER OF PAGES

18. PRICE