DEVELOPMENT OF A GENERAL MODEL TO PREDICT THE RATE OF RADIONUCLIDE RELEASE (SOURCE TERM) FROM A LOW-LEVEL WASTE SHALLOW LAND BURIAL FACILITY

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ABSTRACT

Federal Code of Regulations 10 CFR 61 requires that any near surface disposal site be capable of being characterized, analyzed, and modeled. The objective of this program is to assist NRC in developing the ability to model a disposal site that conforms to these regulations. In particular, a general computer model capable of predicting the quantity and rate of radionuclide release from a shallow land burial trench, i.e., the source term, is being developed. The framework for this general model has been developed and consists of four basic compartments that represent the major processes that influence release. These compartments are: water flow, container degradation, release from the waste packages, and radionuclide transport. Models for water flow and radionuclide transport rely on the use of the computer codes FEMWATER and FEMWASTE. These codes are generally regarded as being state-of-the-art and required little modification for their application to this project. Models for container degradation and release from waste packages have been specifically developed for this project. This paper provides a brief description of the models being used in the source term project and examples of their use over a range of potential conditions.

INTRODUCTION

Before the issuance of the Nuclear Regulatory Commission (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 10 CFR 61, the NRC Technical Position on Waste Form, and the NRC Technical Position on Site Suitability, Selection and Characterization. One specific regulation in 10 CFR 61 requires that any near surface disposal site be capable of being characterized, analyzed, and modeled.

The objective of this program is to assist NRC in developing the ability to model a disposal site that conforms to the regulations in 10 CFR 61. 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.

The results of this modeling work will 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, determinations 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 radioactivity from a trench and burial site.

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

The task of modeling the performance of a low-level waste disposal facility can be divided into four stages: a) conceptualization, this involves identification of the system and of the important physical and chemical processes to be modeled and developing a framework for model development; b) quantification, which requires translating the conceptual model into a mathematical model capable of predicting system performance; c) application, use of the mathematical model to predict the behavior of the system over a range of conditions; and d) validation, comparison of the model predictions to experimental data.

Currently, this project has been most heavily involved with the first three stages of model development. The next sections of this paper will describe the status of work on stages a) through c). This will be followed by a final section which presents conclusions and describes future work.

MODEL CONCEPTUALIZATION

NRC is interested in developing the capability to predict the rate of radionuclide release from low-level waste disposal sites. These sites include shallow land burial as well as alternative disposal techniques that are being proposed.

Currently in the United States, all low-level waste disposal sites have used shallow land burial. Therefore, initial modeling efforts have been structured towards calculating radionuclide release from a shallow land disposal trench.

A shallow land burial site is a complicated physicochemical system containing different types of soil (cover, water infiltration cap, backfill in the trench), different types of waste containers (carbon steel drums, high integrity containers,...) , and different types of waste forms (compacted lab trash, concrete solidified wastes, activated metals,...), with different chemical composition due to the different waste streams (filter sludges, evaporator bottoms, ion exchange resins,...).

A complete model of all of the physical and chemical mechanisms that influence release is not possible due to the complexity of a shallow land disposal site. Thus, conceptualization of the system requires simplifying assumptions. These assumptions are based on identifying mechanisms that influence release; deciding which mechanisms have the greatest influence on release; and maintaining a balance between over simplification which tends to be unrealistic and keeping the system from requiring development of models which currently can not be supported due to insufficient knowledge or data. A primary objective during the conceptualization stage of this program was to make the solution procedure flexible enough to allow incorporation of new models to represent different alternative disposal methods while retaining the basic procedures used for modeling shallow land burial. This was accomplished by structuring the computer code to consist of a series of modules that represent the major physical processes that influence disposal site performance.

In this paper, 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 as: a) water infiltration which brings leachant to the container; b) container degradation, which upon container breach allows leachant to contact the waste form; c) re-lease from waste packages which supplies radionuclides to the water, and d) radionuclide transport from the waste form to the edge of the disposal unit (1). These four processes form the framework for modeling low-level disposal sites. Within each of these four modules, there may be several models to represent different materials and/or different phenomena that influence release. For example, within the waste form leaching module, there could be different models to represent leaching from different types of waste forms.

The next section discusses the translation of these four major physical processes into mathematical models capable of predicting performance. Currently, each of the four processes has been modeled and tested individually. Coupling of the models into a unified system model remains to be done.

MODEL SELECTION/DEVELOPMENT

Modeling the processes leading to radionuclide release from a disposal trench is a complex procedure. To avoid duplication of effort, whenever possible, existing models were selected for use in this project. Existing models have been selected in the areas of water flow and radionuclide trans-port. Models for container degradation and release from the waste form have been specifically developed for this project.

Modeling Water Flow and Radionuclide Transport

Water flow and radionuclide transport are intimately connected because movement of water is expected to be the most dominant mechanism of trans-porting radionuclides through the disposal trench. Thus, both flow and transport are discussed in this section.

In predicting water flow in an unsaturated porous medium, i.e, a shallow land burial facility, 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 waste forms, make closed-form analytical solutions impossible to obtain. Therefore, numerical solution techniques are needed to solve the mass balance equation.

The necessity of predicting water flow in an unsaturated porous medium has been recognized by the NRC and others interested in the safe disposal of hazardous and radioactive waste. Both NRC and Electric Power Research Institute have had contractors conduct reviews to evaluate the computer codes available for predicting water flow and/or solute transport (2,3). These reviews resulted in identification of a number of state-of-the-art computer codes that are well documented, and available to the public.

Based on these reviews, the computer codes FEMWATER and FEMWASTE were selected in our work to model water flow and radionuclide transport in the unsaturated zone (4,5). FEMWATER predicts water flow and provides the flow velocities used by FEMWASTE in calculating radionuclide transport. The FEMWATER/FEMWASTE computer codes are finite element codes that can model the time dependent evolution of water flow/waste transport in two spatial dimensions. They can be used in conjunction or independently. The primary reason for selecting these codes over other state-of-the-art codes was that NRC has used them and gained some familiarity in the use of these codes (6,7).

Modeling Container Degradation

Before water can contact a waste form and thereby release radioactivity to solution, the container must be breached. Therefore, to predict release it is critical to know the time at which breach occurs for each container, the number of breached containers at any given time, and the area breached per container.

Because of low cost and relative durability, carbon steel containers are the most commonly used low-level waste package containers. These containers are subject to chemical attack (corrosion) which will eventually lead to breach. Carbon steel containers are susceptible to general as well as localized corrosion in soil environments. Since localized corrosion could lead to exposure of the wastes to leachant at earlier times than generalized corrosion, an empirical model of localized corrosion has been developed (8).

In this model, as a first approximation, localized corrosion as represented by pitting corrosion was assumed to be the major degradation mechanism for carbon steel containers. The central points which need to be addressed for modeling pitting corrosion are the following: a) the kinetics of pit growth in terms of depth and radius; b) the number of pits per unit area on the sample as a function of time; and c) the distribution of pit depths.

In the present study, attention is focused on metallic corrosion in soil environments with emphasis on the development of a relationship between maximum pit depth and time as a function of pH, soil resistivity, and degree of soil aeration.

The principal source of soil corrosion data is a report from the National Bureau of Standards (NBS) (9). The NBS study considered ferrous samples in 125 different soils characterized by a wide range of resisitivities (64-45,000 ohm-cm), pH values (3.1 9.8), moisture contents (2.8 57.8%), and soil aeration properties over a time period of 18 years. In the NBS study, it was found that the maximum pit depth, h, due to pitting corrosion was closely represented by the relationship:

$$h = kt^{n} \qquad (1)$$

where t is the exposure time of the sample in years, k is the pitting parameter in mils/(years)^{n}, and $n > 0$ is a parameter related to the degree of soil aeration. Values for n were obtained in the NBS study; it was found that the better the degree of aeration, the lower value of n. The values of n ranged from 0.26 for the most aerated soils to 0.59 for the least aerated soils.

The central objective of the present investigation is the determination of the dependence of the pitting parameter k on the soil properties using the NBS data on corrosion in soils. The result of a detailed linear correlation analysis of k versus pH and soil resistivity revealed that k is principally influenced by the pH of the soil. The resistivity of the soils plays a minor role. Based on the linear regresssion analysis, the following two relationships were deduced:

$$k = 5.74\,(9.9 - \mathrm{pH}), \qquad (2)$$

$$k = 5.05\,(2\mathrm{pH} - 10.3), \qquad (3)$$

for acidic and alkaline soils, respectively. The two linear relationships intersect at the point specified by $\mathrm{pH} = 6.9$ and $k = 17.3$.

With Eqs. (2) and (3) plus values for n deduced from the NBS data, an estimate for maximum pit depth can be obtained. This can be compared to the thickness of the container to determine the time of penetration.

To translate from a single pit breaching a container to the area of the container breached due to pitting requires a number of assumptions. First, it is assumed that the pit continues to grow at the same rate once it has penetrated a container and the penetrated area grows in a circular fashion. These assumptions lead to an estimate of the area exposed by a single pit. Next, given knowledge about the pit depth distribution and the number of pits per unit area, an estimate of the number of pits per unit area that grow at a rate fast enough to penetrate the container can be made. The product of the number of penetrating pits per unit area and the area exposed by a single pit gives the area breached due to pitting.

Modeling Release From Waste Packages

After the container has been breached, water can contact the waste form and cause the release of radionuclides. There are many different waste forms and waste streams being disposed of in shallow land burial facilities and each may have different release characteristics. Thus, model development has focused on developing a generic model with broad applicability (10).

Specifically, the leaching model that is under development is directed towards radionuclide release from porous waste forms that are disposed of in corrodible outer containers. Two broad categories of low-level radioactive wastes may be modeled by this approach:

(1) Class A, B, and C wastes that have been solidified in concrete or another porous solid material.

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

Two main processes have been modeled for leaching. In the first, called the rinse model, water flows through the breached area of the outer container into the waste by matric suction until all of the available pore or void space of the waste package (and, for monolithic wastes, of the waste form itself) is filled. Once the pore and void spaces of the waste are occupied by water and solubilization of radionuclides has taken place, the process of exit of this water (now containing radionuclides), may begin. The increment of water taken into the container is, for that time interval, to be a replacement of an equivalent volume of water exiting the waste laden with leached radionuclides.

The second leaching model being developed, the diffusion model, involves the approximation of filled, or partially-filled pores in the waste as extended sources, initially homogeneous in concentration, whose "length" is determined by the pore "length". Radionuclides are allowed to diffuse through the extended source region, the pore, to the edge of the waste form and into a clean water column (uncontaminated leachant volume). The clean water column length will be proportional to water flow.

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, 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.

Among the quantitative factors required for this model are: the water flow rate, the area breached on the outer container, and the initial distribution of species within the waste form, that is the fractional allocation of the species under consideration that reside on the surface, inner pore surfaces, and in the waste matrix. From the water flow rate and the breached area, the volume of water flowing into the waste form region per unit time and the total volume that has entered the container can be calculated. Using this information and the species distribution in the waste form, the incremental and cumulative fractional release can be calculated. The incremental fractional release can be used as input to the transport calculation as the source of radionuclides.

MODEL APPLICATIONS

Water Flow in Shallow Burial Trenches

Water flow has been modeled for a vertical cross-section perpendicular to the longitudinal axis of a generic low-level waste burial trench (11) using the computer code FEMWATER-1 (4). The trench is situated in the unsaturated zone and is approximately 28 meters wide and 10 meters deep, and is composed of three types of soil: a relatively high hydraulic conductivity gravel cap on top; a relatively low hydraulic conductivity clay layer beneath the cap; and soil backfilled into the waste storage region. Surrounding the trench is a region of undisturbed soil.

Different steady-state water flow problems have been simulated by varying the amount of rainfall that enters the ground, the width of the gravel and clay layers relative to the width of the waste region, and hydraulic proper-ties within the trench, that is, waste containers were simulated as low hydraulic conductivity regions as compared to the backfill.

Based on the results of these simulations, the following conclusions can be drawn: 1) Steady-state simulations show that the low-conductivity clay layer is effective in preventing rainwater from infiltrating directly into the trench region. However, water in the gravel cap flows towards the edge of the trench, and enters the trench from the side due to capillary suction. There-fore, the moisture barrier must be extended beyond the sides of the trench to significantly reduce water flow through the trench bottom. For the trench geometry and material properties modeled, a 42% reduction in overall flow through the trench bottom was achieved by an extension of the trench cap 8 meters beyond the edge of the trench (this is approximately 25% of the width of the trench); and, 2) In our simulation, having 25% of the area filled with low-conductivity waste packages did not significantly influence the net water flow through the trench. However, increased water flow occurs between the waste packages, resulting in regions of relatively high water velocity and regions of lower water velocity.

Container Degradation

The pitting corrosion model described briefly in an earlier section of this paper and more fully in reference (13) has been applied to determine the area breached due to pitting as a function of time for a range of pH and soil aeration conditions. For these calculations, it was assumed there was a fixed number of pits penetrating the carbon steel drum wall thickness of 0.127 cm, 50 mil, and the kinetics of pit growth were described by Eqs. (1) (3). The number of penetrating pits was estimated based on experimental data. Due to uncertainties in the applicability of the assumptions and the experimental data, the magnitude of the predicted breached area may not be accurate. However, the following trends are apparent: a) The degree of soil aeration, which determines the value of the exponent in Eq. (1), is the most important factor in determining the amount of pitting. Poorly aerated soils exhibit much greater pitting than well-aerated soils; b) Soil pH is also an important factor in pitting. Soils with near neutral pH show the least pitting; and, c) The time at which the first pit penetrates will be brief, less than 2 years for the range of conditions modeled.

Release From Waste Packages

The rinse model has been applied over a range of water flow rates, pitting rates, and pore/void space fractions in the waste. The initial release time and period-of-release curves for this model were found to depend very heavily on water flow rates and pore/void space fractions. For example, a 10% void fraction waste would yield first release in year 6 for a water flow rate of 100 cm/year, while for water flow of 10 cm/year the initial release would occur about year 13, and for 1 cm/year, year 28. These times correspond to that needed to accumulate the void space volume of water. The variation in first release in not linear with void space fraction. For example, given all other parameters equal, a 20% void space waste would have initial release times of 8, 16, and 31 years, respectively, for the water flow rates given above (1,10).

Also, the period-of-release or length of leaching for the sets of conditions outlined above could range from 4-5 years (100 cm/year water flow rate), through 10-14 years (10 cm/year water flow rate), to 27-59 years (1 cm/year water flow rate). These are all results for release of a surface-residing species in the waste; they do not include the solid-phase diffusive contributions expected to come into play at long times (given the low diffusion coefficient values expected for solid-phase diffusion) (13).

The diffusion model results show, as would be expected, a very strong dependence on the value of the diffusion coefficient. A range of values has been studied corresponding to those for ions in aqueous solutions through "effective" diffusion coefficient values reported for radionuclide ions released during leaching tests under saturated conditions.

The diffusion model results also indicate that the magnitude of the clean water/leachant volume available does not have a significant effect on the amount of diffusive release for early times (times before complete homogenization of the pore water-leachant system). It appears that short-term diffusive releases may best be modeled to occur during intermediate/low water flow while the rinse release model may better accommodate circumstances in which water flow is high. Implicit in extending the results of either model for any length of time is that the process is uninterrupted; i.e., calculation of the diffusion release for 100 years implies that diffusion is allowed to proceed for 100 years. This may be unrealistic given the fluctuating nature of water flow in the trench. The coupling of the rinse and diffusion models is planned to accommodate these circumstances; rinse release can be modeled for a period followed by diffusive release and then back to rinse release as needed to reflect changing water flow conditions. Each type of release model has been developed as an individual module, each leads to incremented depletion of the total available waste package source term, and the modules can easily be interfaced in sequence.

Radionuclide Transport

Simulations of radionuclide transport from a generic shallow land burial trench have been performed for a range of water flow rates, dispersivity values, and distribution coefficients using the computer code FEMWASTE-1 (7). The system modeled is geometrically identical to the system described earlier for water flow. That is, the trench is composed three regions, a gravel layer on top, a moisture migration barrier of clay beneath the gravel, and a region containing backfill and waste. Water flow rates and soil moisture content were provided through use of the companion code FEMWASTE-1 (8,13).

This study had two objectives. The first was to determine the relative importance of advection, dispersion, diffusion, and sorption over a range of expected conditions by conducting a number of numerical simulations in which the transport parameters were varied. Second, any gaps in the data necessary for accurate prediction of radionuclide transport were to be identified.

For the modeling assumptions used and the range of parameters tested, the water flow velocity plays the major role in redistributing radionuclides within the trench except in the case of unexpectedly high values of dispersivity. Dispersion was always found to play a significant role in determining transport. This was particularly apparent in the clay and gravel layers above the waste region which are upstream from the source and therefore, radio-nuclides are transported to these regions only by the dispersion mechanism. Sorption decreased the magnitude of the radionuclide concentration and flux, and had the apparent effect of reducing the velocity with which the radionuclides were transported. Diffusion was found to be unimportant in determining radionuclide transport.

Two parameters were identified in which there is a shortage of relevant modeling data; these were dispersivity and radionuclide sorption in unsaturated soils. Of the two, sorption is more likely to have a greater influence on transport.

CONCLUSIONS

The framework for a generic model capable of predicting the release of radionuclides from a low-level waste disposal trench located in the unsaturated zone has been developed. This framework consists of four basic compartments: water flow, container degradation, waste form leaching, and radionuclide transport. Models for water flow and radionuclide transport rely on the use of the computer codes FEMWATER and FEMWASTE. These codes are generally regarded as being state-of-the-art and required little modification for their application to this project. Models for container degradation and waste form leaching were specifically developed for this project. Testing of the models to determine important processes influencing release of radio-nuclides from the trench has been done under a wide range of conditions.

Currently, modeling of container degradation and waste form leaching has focused on the materials believed to be the most widely used in shallow land disposal (carbon steel containers and porous waste forms). To check the validity of this assumption and to determine where additional work needs to be done in defining the initial amount of radioactivity and container types disposed of in a waste trench, the next phase for this project will involve a detailed characterization of the disposal practices at the existing disposal sites. Based on this work, recommendations will be made as to future modeling work in this project.

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REFERENCES

1. T. M. Sullivan and C. R. Kempf, "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," NUREG/CR-4897, BNL-NUREG-52066, (1987).
2. C. A., Oster, "Review of Ground-Water Flow and Transport Models in the Unsaturated Zone," NUREG/CR-2917, PNL-4427 (1982).
3. C. T. Kincaid, J. R. Morrey, and J. E. Rogers, "Geohydrochemical Models for Solute Migration. Volume 1: Process Description and Computer Code Selection," EPRI EA-3417, Vol. 1, (1984).
4. G. T. Yeh, "FEMWATER: A Finite-Element Model of Water flow Through Saturated-Unsaturated Porous Media First Revision," ORNL-5567/R1 (1987).
5. G. T. Yeh and D. S. Ward, "FEMWASTE: A Finite-Element Model of WASTE Transport through Saturated-Unsaturated Porous Media," ORNL-5601 (1981).
6. G. T. Yeh, "Training Course No. 1: The Implementation of FEMWATER Computer Program," NUREG/CR-2705, ORNL/TM-8327 (1982).
7. G. T. Yeh, "Training Course No. 2: The Implementation of FEMWASTE (Finite Element Model of WASTE) Computer Program," NUREG/CR-2706, ORNL/TM-8328, (1982).
8. S. M. Mughabghab, and T. M. Sullivan, "Investigation of the Pitting Corrosion of Low-Carbon Steel Containers," Waste Management '88, (1988).
9. M. Romanoff, Underground Corrosion, National Bureau of Standards Circular 579 (1957).
10. C. R. Kempf, "A Mechanistic Model for Leaching from Low-Level Radioactive Waste Packages," Waste Management '88, (1988).
11. C. J. Suen, "Modeling the Flow of Water in and around Shallow Burial Trenches," Waste Management '88, (1988).
12. T. M. Sullivan, "Prediction of the Migration of Radionuclides to the Boundary of a Shallow Land Burial Trench," Waste Management '88, (1988).
13. C. R. Kempf, C. J. Suen, S. Mughabghab, and T. M. Sullivan, "Low-Level Waste Source Term Evaluation, Quarterly Progress Report, January March, 1987,", Brookhaven National Laboratory, WM-3276-2, (1987).