Conf-920307--27

EGG-M--91452

DE92 017944

Evaluation of EPICOR-II Resin/Liner Lysimeter Investigation Data Using "MIXBATH," a One-Dimensional Transport Code[a,b]

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a. Work supported by the U.S. Nuclear Regulatory Commission, Office of Nuclear Regulatory Research, under DOE Idaho Field Office Contract DE-AC07-76ID01570.

b. The research was performed under appointment to the Environmental Restoration and Waste Management Fellowship program administered by Oak Ridge Associated Universities for the U.S. Department of Energy.

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ABSTRACT

The computer code MIXBATH has been applied to compare model predictions with six years of leachate collection data from five lysimeters located at Oak Ridge and five located at Argonne National Laboratories. The goal of this study was to critique the applicability of these data for use as a basis for the long-term prediction of release and transport of radionuclides contained in Portland type I-II cement and Dow vinyl ester-styrene waste forms loaded with EPICOR-II prefilter ion exchange resins. MIXBATH was useful in providing insight into information needs for long-term performance assessment. In most cases, the total activity released from the lysimeters over the test period was indistinguishable from background, indicating a need for longer-term data collection. In cases where there was both sufficient information available and activity released, MIXBATH was able to predict releases within an order of magnitude of those measured. Releases are extremely sensitive to the soil partition coefficient and waste form diffusion coefficient, and these were identified as the key data needs for long-term performance assessment.

DISCLAIMER

INTRODUCTION

A portion of the EPICOR-II prefilters, expended in the cleanup of Three Mile Island Unit-2 (TMI-2) liquid waste, was subject to examination in the EPICOR-II Research and Disposition Program initially funded by the Department of Energy (DOE). In FY 1984, this project was transferred to the U.S. Nuclear Regulatory Commission (NRC) as the Low Level Waste Data Base Development TMI-2 EPICOR-II Resin/Liner Investigation. Among the tasks initiated in that program was a field testing project to address different aspects of the disposal of prefilters and their contents.

Field testing of waste forms containing EPICOR-II ion exchange resins using lysimeters is a unique project marking the first time such a highly loaded ion exchange resin (up to 74.7 TBq/m^3) had been incorporated into commercial formulations of solidified media for use in waste-form testing. This project, to test waste form performance in soil systems, as measured by quantity of released material, began generating data in 1986.

Modeling, analysis, and monitoring requirements of waste disposal site conditions are implicit in Title 10 of the Code of Federal Regulations Part 61 (10 CFR 61), "Licensing requirements for Land Disposal of Radioactive Waste." This regulation has stimulated our study on the applicability of EPICOR-II lysimeter data to transport modeling and performance assessment.

METHODS AND MATERIALS

Lysimeters and Waste Forms

There are two arrays of five lysimeters used in the Low Level Waste Data Base Development TMI-2 EPICOR-II Resin/Liner Investigation. Five lysimeters are located at Argonne National Laboratory (ANL-E), and five are located at Oak Ridge National Laboratory (ORNL). The lysimeters are right cylinders (0.91 m ID by 3.12 m in height). This size allows for unrestricted flow of soil water, sufficient depth to prevent freezing of the waste forms, and sufficient waste form shielding to prevent unnecessary radiation exposure to experimenters working above the waste. The lysimeters are each equipped with five porous cup soil water

samplers, soil moisture/temperature probes complete with an automated data acquisition system, and a leachate collection system.

Four lysimeters at each site are filled with soil; the remaining one is a control lysimeter filled with an inert material, Unimin silica oxide sand. Two different soils are used. ANL-E lysimeters use a local indigenous soil that fits the NRC criterion for a representative Midwestern type. It is a Morley silt loam with the surface layer removed. The resulting subsurface soil is a clay loam. ORNL lysimeters use a soil that is intended to approximate the soil found at Barnwell, South Carolina. It is soil from the C horizon of a Fuquay sandy loam obtained from the Savannah River Plant (SRP) adjacent to the Barnwell facility in South Carolina. This soil is texturally similar to the soil found at Barnwell.

The waste forms used in the field test are composed of EPICOR-II resin wastes. Two resin types were used in the solidification project. The first type was a mixture of synthetic organic ion exchange resins (phenolic cation, strong acid cation, and strong base anion resins), while the second type was a mixture of synthetic organic ion exchange resins (strong acid cation and strong base anion resins) with an inorganic zeolite. Portland type I-II cement and vinyl ester-styrene (VES) (a proprietary solidification agent developed and supplied by the Dow Chemical Company) were used to solidify both types of resin wastes. Waste stream characterization identified Cs-134, Cs-137, Sr-90, and Co-60 as the radionuclides present. Individual cylindrical waste forms were produced with an average diameter of 0.048 m and a height of 0.076 m. Seven waste forms were stacked, one on top of the other, in each lysimeter. Fig. 1 is a diagram of a lysimeter with the waste forms installed. Table I specifies the type of waste forms that were placed in each lysimeter. Leachate is removed from moisture cups 1 through 5 and the leachate container (see Fig. 1). The leachate is analyzed for gamma-emitting radionuclides and Sr-90. A complete description of the lysimeters and waste forms is given in NUREG/CR-4498 (1).

Use of the MIXBATH Code

The computer code MIXBATH (see NUREG/CR-5681) (2) is in the public domain and may be obtained from Dr. T. M. Sullivan of Brookhaven National Laboratory, Upton, NY 11973. MIXBATH is used to model the release of radionuclides from a waste form in a failed container surrounded by a porous medium containing a solute. The solute is treated as a well-stirred fluid, i.e., a mixing bath. The solute concentration is calculated using a mass balance that depends on

the flow rate, the amount of partitioning between the porous medium and solute, the size of the mixing bath, the radioactive decay rate, and the rate of release from the waste form. The waste form is modeled using a one-dimensional finite difference model. Releases from the waste form are directly coupled to the solute concentration. MIXBATH has the capability to simultaneously consider three waste form release mechanisms: diffusion, dissolution, and surface rinse limited by partitioning. A complete mathematical description of the model used in MIXBATH is given in NUREG/CR-5681.

Available Data, Estimates, and Assumptions

In this study, we modeled the release of Cs-137 and Sr-90 from the waste forms. We considered diffusion from a cylinder to be the most appropriate release process for both the Portland type I-II and VES waste forms. This is consistent with Cs-137 results from American Nuclear Society (ANS) 16.1 (3) leach tests performed on the waste forms created in this project (4). The waste form diffusion coefficients for Cs-137 were computed from those results while those for Sr-90 were estimated based on measurements of similar waste forms of equal size (5).

The mass balance for the solute concentration requires the volumetric flow rate per unit area, i.e., the Darcy velocity. This was estimated from the leachate analysis data obtained in 1989 (6). Soil/water partition coefficient (Kd) values were also needed but were not available for these soils. These were estimated from a previous work published by Looney (7), which was reproduced in NUREG/CR-5387 (8). Tables II, III, and IV list the values used in this study for the most important parameters. These include the soil/water partition coefficients, the decay constants, the diffusion coefficients (D) for each waste form and isotope, and the Darcy velocities. It should be noted that the values for the diffusion coefficients shown in Table III for VES have a range of variability between four and six orders of magnitude.

Those values are laboratory determined and give an indication of how these data can vary. The Portland cement diffusion coefficients shown are within one order of magnitude. This highlights the necessity of using lysimeter-specific parametric values.

A number of preliminary calculations were completed using the MIXBATH model with the data and parameters described above. The output of this model is the concentration of Cs-137

Table II. Partition coefficients (cm^3/g) of three soils used in lysimeters.

Radionuclide	Value Used	Model Parameters: Lower Boundary	Model Parameters: Upper Boundary
	Morley Silt Loam		
Cs-137	10^3	10^1	10^5
Sr-90	$10^{0.9}$	10^0	10^3
	C Horizon of Fuquay Sandy Loam		
Cs-137	10^3	10^1	10^5
Sr-90	$10^{0.9}$	10^0	10^3
	Unimin Silica Oxide Sand (Inert Material)		
Cs-137	0[a]	10^1	10^5
Sr-90	0[a]	10^0	10^3

a. The value assumed for essentially inert material.

Decay Constants (s^{-1})	
Cs-137	7.28×10^{-10}
Sr-90	7.57×10^{-10}

Table III. Diffusion coefficients of waste forms used in lysimeters (cm^2/s).

Waste Form	Radionuclide: Cs-137[a]	Radionuclide: Sr-90[a]	Radionuclide: Sr-90[b]
Vinyl ester-styrene	3.30×10^{-14}	2.0×10^{-10}	1.35×10^{-8}
Portland type I-II cement	5.0×10^{-11}	9.6×10^{-10}	4.0×10^{-10}

a. See Reference (4).

b. See Reference (5).

Table IV. Darcy velocities of soils used in lysimeters.

Lysimeter Number	Darcy Velocity (cm^3/s) ANL	ORNL
1	9.42×10^{-7}	3.07×10^{-6}
2	1.10×10^{-6}	3.10×10^{-6}
3	1.65×10^{-6}	3.12×10^{-6}
4	1.34×10^{-6}	3.16×10^{-6}
5	2.59×10^{-6}	3.60×10^{-6}

and Sr-90 along with the cumulative release of these radionuclides. These predicted values were compared to the experimentally measured values.

RESULTS AND DISCUSSION

Results of this preliminary lysimeter performance assessment fell into two categories, those in which the prediction and measured values were in poor agreement, and those in which the parametric information available was broad enough for accurate modeling predictions. Unavoidably, the difference was caused by a lack of waste-form-specific diffusion coefficient data. Prediction accuracy was a function of the estimated soil/water partition coefficient. Also, in almost all instances, the activity in the leachate recovered from the lysimeters was essentially at background levels, and because of the uncertainty associated with such a small activity, actual model validation was not possible.

Fig. 2 shows plots of predicted and measured Sr-90 cumulative activity versus time for a lysimeter located at ORNL. Two predictions are shown. One prediction uses a diffusion coefficient of 4.0×10^{-10} cm^2/s, the other a value of 5.0×10^{-11} cm^2/s. If one assumes a waste-form-specific Sr diffusion coefficient equal to that of Cs (5.0×10^{-11} cm^2/s), then the MIXBATH prediction and measured values agree fairly well as seen in Fig. 2. The use of such a value for Sr-90 has not been verified experimentally, but it is consistent with releases measured during lysimeter experiments using these waste forms (6) and results in reasonable prediction. The waste

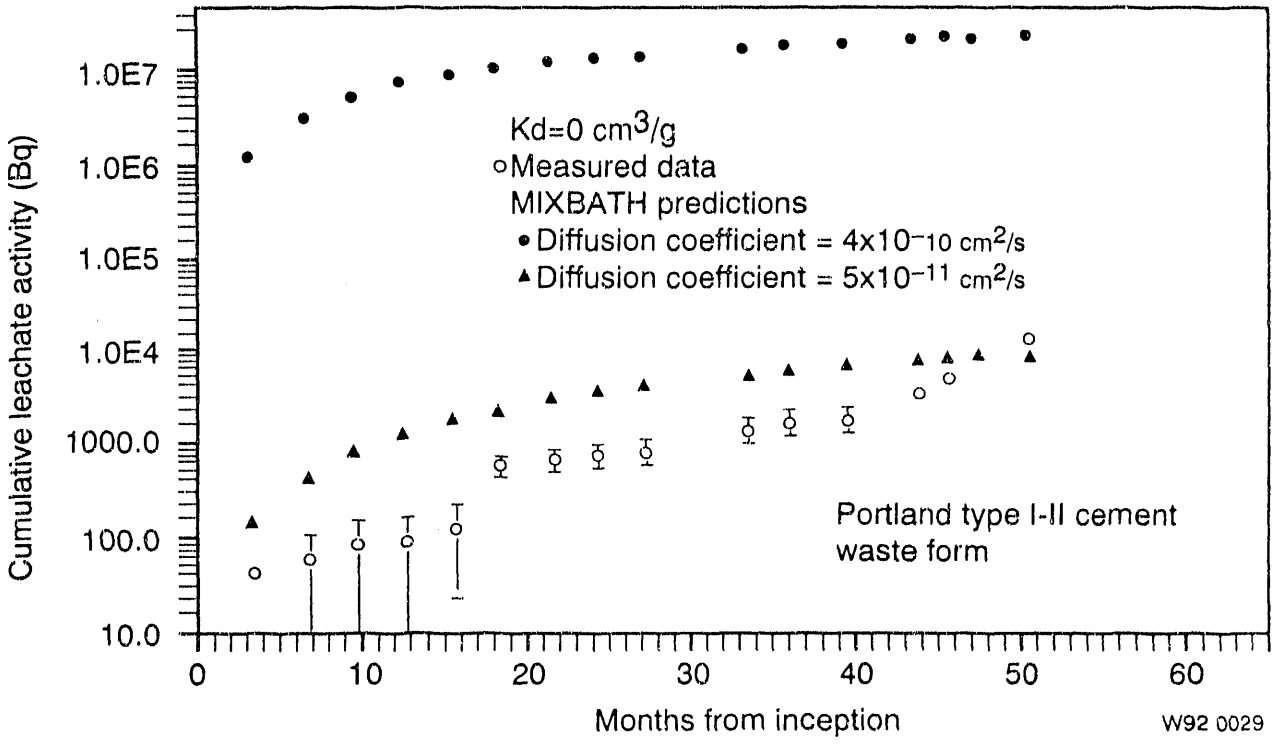

Fig. 2. Comparison of Sr-90 cumulative activities for measured data from ORNL lysimeter No. 5 with MIXBATH predicted results.

form diffusion coefficient of 4.0×10^{-10} cm^2/s has been measured in laboratory leach tests for Portland cement waste forms that do not contain ion exchange resin beads. The actual value of the EPICOR-II waste-form-specific diffusion coefficient (see Table III) is the value of 4.0×10^{-10} cm^2/s used here. The curve generated by using MIXBATH and applying a waste form diffusion coefficient of 4.0×10^{-10} cm^2/s is also shown in Fig. 2. Use of this value produces predictions that are five orders of magnitude greater than measured values. This demonstrates the necessity of obtaining waste-form-specific diffusion coefficients.

The cumulative activity obtained from the lysimeters is too small at this time for code validation. However, there are similarities between the predicted and measured curves. The plots of Portland cement shown in Fig. 2 are thought to be typical of Sr-90 release in both VES and Portland type I-II cement waste forms. Sr-90 diffusion coefficients used for prediction of cement waste form behavior could be different than actual values from lysimeters. Experience in the lysimeter experiment of Reference (6) indicates that for Cs and Sr, diffusion coefficients are probably of the same order of magnitude for Portland cement and VES.

Another possible reason for deviation between measured and predicted values is the estimate of the partition coefficient (Kd). Fig. 3 demonstrates the possible range of predicted values that could be obtained using MIXBATH when only the potential variability of the partition coefficient is considered. This figure shows the range of predicted values if Kd is set to both 1.0 and 1000 cm^3/g when plotted against time for a hypothetical lysimeter in which the Sr-90 leachate activity is of interest. This emphasizes the sensitivity of code predictions (such as those of MIXBATH) to modeling parameters.

An example in which the MIXBATH prediction is reasonably accurate is shown in Fig. 4, which indicates how the measured value of the diffusion coefficient and close approximations of the partition coefficient (Table II) can significantly increase the accuracy of the prediction. Although this prediction is within one order of magnitude, it engulfs all of the uncertainty associated with estimation of the partition coefficient and subsequent retardation coefficient.

Two items that should be determined experimentally are the soil/water partition coefficient and the waste-form-specific diffusion coefficient. At early times, when the fractional release is low, releases from MIXBATH approximately follow the inverse of the soil/water retardation coefficient. Similarly, releases from the waste form are approximately proportional to the square root of the waste-form-specific diffusion coefficient at early times.

The small quantity of activity present in leachate samples, relative to the total waste form loading, indicates that the main body of activity has not reached the bottom of these lysimeters and could require years to do so. This is based on the comparison of the small leachate activities measured in the soil-filled lysimeters with the relatively high leachate activities measured in control lysimeters filled with sand, an essentially inert material.

MIXBATH worked well for the purpose of this preliminary performance assessment. It demonstrated the areas of data that will need to be expanded prior to a complete performance assessment and the embryonic nature of the data presently available. We feel MIXBATH has been a useful code for application in this situation. MIXBATH is limited to advection-dominated situations for which there is no interest in transient flow conditions. It was appropriate at this level of analysis in which the effects of dispersion are thought to be insignificant and the values for average flow rate were readily available. Dispersivity values and soil hydraulic properties were not measured experimental parameters within the lysimeter data set. These values will be

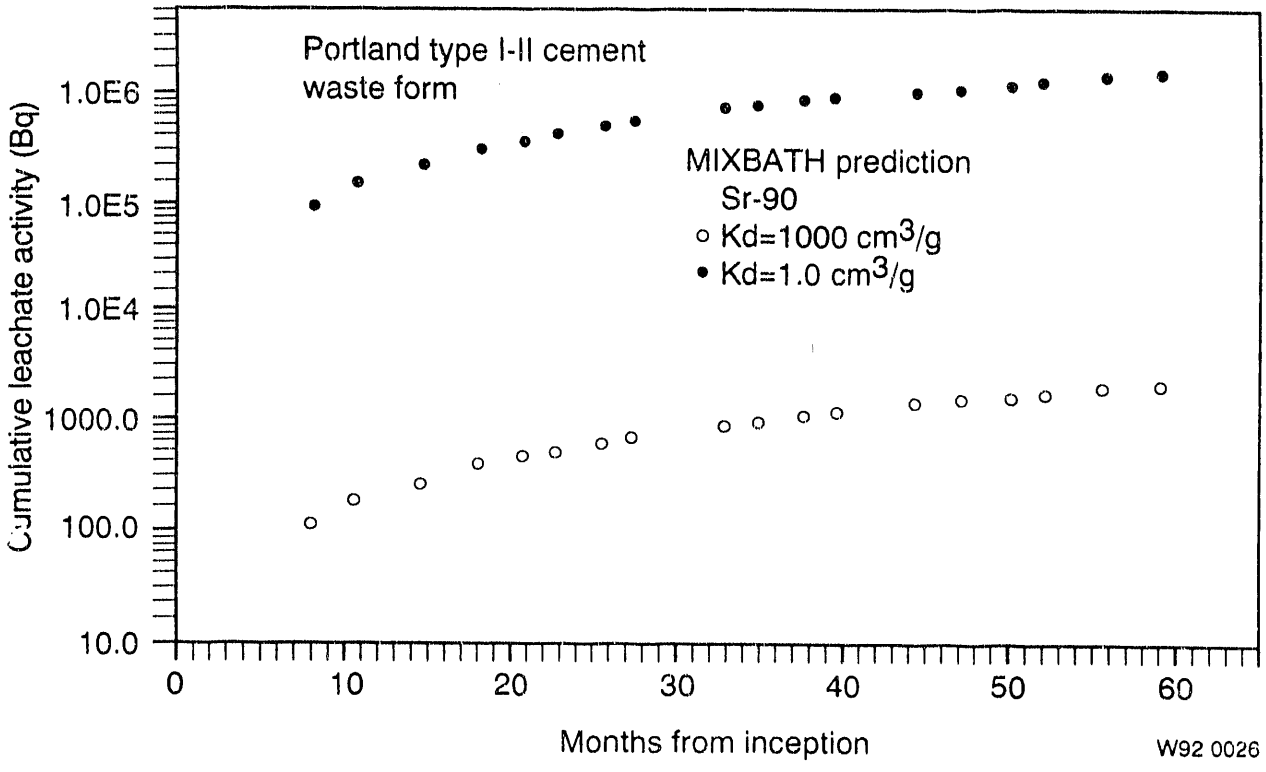

Fig. 3. Effect of Kd variation on cumulative predicted activity for Sr-90 in a hypothetical lysimeter.

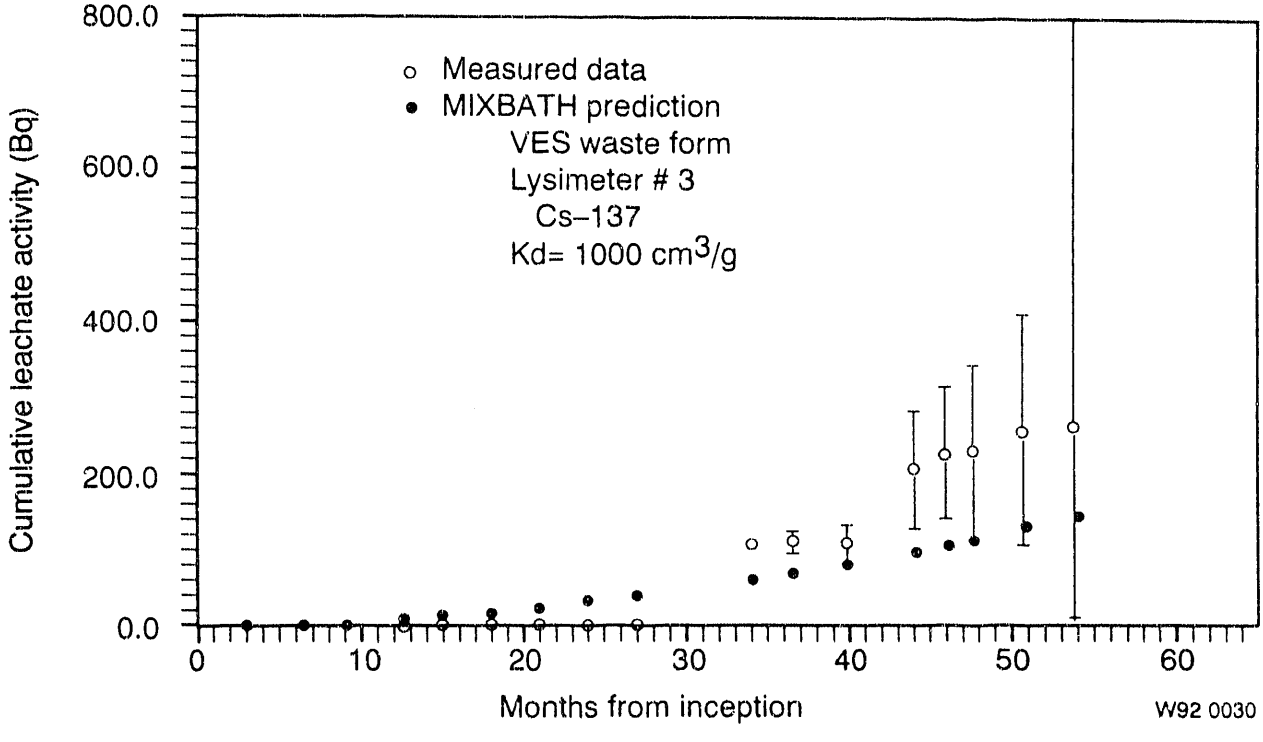

Fig. 4. Comparison of Cs-137 cumulative activity for measured data at ORNL lysimeter No. 3 with MIXBATH prediction.

necessary in the future if detailed modeling studies are performed. Absence of these data further justifies use of a "simplified" model like MIXBATH.

In all cases studied, the activity of the release measured in lysimeter leachate samples has been a small fraction (less than 0.01%) of the total inventory, and it is not yet possible to determine if long-term trends have been established. Several more years of data collection are necessary for definitive performance assessment of the waste forms being studied.

CONCLUSIONS

1. To limit model/prediction uncertainties, several parameters need to be measured in the laboratory. The most important include the soil/water partition coefficients and waste-form-specific diffusion coefficients. For more detailed modeling, soil flow parameters (e.g., hydraulic conductivity as a function of moisture content, and the longitudinal and transverse dispersivities) need to be known.

2. In lysimeters with experimentally determined diffusion coefficients, in which there were high enough leachate concentrations of Cs-137 for comparison between predicted and experimental results, MIXBATH was able to predict the observed leachate concentrations of Cs-137 within one order of magnitude. MIXBATH worked well as a first approximation during performance assessment of the EPICOR-II waste forms. MIXBATH was also successful in estimating releases of Sr-90 from EPICOR-II waste forms using estimated diffusion coefficient values for Sr-90.

3. Uncertainties in present MIXBATH predictions are mainly a function of the uncertainty in estimates of soil/water partition coefficients.

4. Not enough long-term EPICOR-II lysimeter data exists on which to base the validity of transport models. Several more years of data collection will be necessary to accomplish this task.

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