Waste Isolation Safety Assessment Program

NUMERICAL ANALYSIS OF RADIONUCLIDE MOVEMENT FROM
A HYPOTHETICAL NUCLEAR WASTE REPOSITORY

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INTRODUCTION

As part of the National Waste Terminal Storage (NWTs) program, the Waste Isolation Safety Assessment Program (WISAP) is developing methods to assess the safety of geologic formations as nuclear waste repositories. One component of WISAP is concerned with the release consequence analysis. Its purpose is to develop methods for simulating the transport of radionuclides by ground water from the repository through the geosphere and to calculate the potential doses to humans. The purpose of this paper is to present an application of this technology to a hypothetical repository in a reference geologic site. This exercise was done in the following steps:

1. Development of a conceptual model based on available geologic and hydrologic data.
2. Development of a release scenario.
3. Simulations of ground-water flow and contaminant transport.
4. Analysis of the model sensitivity to varying input parameters.

The inventory of nuclear waste was assumed to be spent fuel. The results obtained for the base case indicate that the concentration of the nuclides that reach the discharge point will be below MPC levels for uncontrolled water use after dilution in the receiving river. The results of the parametric study emphasize the sensitivity of the model to the assumed values of retardation and dispersion factors and the complex relationships between these parameters. The effect of varying waste leach rates is being investigated.

CONCEPTUAL AND NUMERICAL MODELS

Reference Repository Site

The location of the conceptual repository (site A in Figure 1) was selected, on the basis of available geologic and hydrologic information, within a bedded salt region. The repository is placed 3000 ft (915 m) below the ground surface in a thick section of interbedded salt and shales bounded above and below by water-bearing formations. The lower unit is a limestone aquifer,

FIGURE 1. Modeled Region with Hydrologic Potentials

500 ft (152 m) thick, with an average hydraulic conductivity equal to 45 ft/yr (4.3×10^{-5} cm/sec). The upper aquifer consists of a combination of several geologic units that are hydrologically connected and is thus considered as a single system. This system is 1000 ft (305 m) thick and has an average hydraulic conductivity equal to 100 ft/yr (9.7×10^{-5} cm/sec).

Flow in the upper aquifer is toward the "S" River located approximately 35 miles (56 km) from the repository site (Figure 1). A cross section parallel to the flow lines is shown in Figure 2. The aquifer fluids in both systems

FIGURE 2. Vertical Section Parallel to Hydraulic Gradient

are essentially saturated brines and their potentials, as used in this study, were corrected to fresh water. The head in the lower aquifer exceeds the head in the upper system. At the location of the repository this head difference is found to be approximately 250 ft (76 m).

The repository was assumed to be rectangular, 10,423 ft (3178 m) long and 8148 ft (2483 m) wide, with 98 disposal rooms on 78.38 ft (23.89 m) spacings. Each disposal room is 10,428 ft (3178 m) long, 20 ft (6 m) high and 20 ft (6 m) wide. The inventory of nuclear waste was assumed to be spent fuel.

Release Scenario

The repository was assumed to be breached by a fault aligned with the repository length so that one entire room of waste canisters was exposed. This fault was assumed to extend from the lower aquifer to the upper aquifer

thereby allowing water to move upward through the repository, leaching the waste canisters and contaminating the upper ground water system. The width of the fracture was assumed to be 5 ft (1.5 m) and its hydraulic conductivity to be approximately ten times that of the lower system, or 500 ft/yr ($4.8 \cdot 10^{-4}$ cm/sec). Using the above parameters in a simple one-dimensional analytical model the flow rate through the fracture was calculated to be 19.5 gpm (1.2 l/sec).

Leach Rate Model

A leach rate model was developed to determine the rate of transport of radioactive material by the water flowing through the fracture into the upper aquifer. The model incorporates the effects of flow rate and temperature on waste leach rates obtained from preliminary experimental data.

Specifically, data from leaching experiments on crushed spent fuel (3 cm^2 of surface area per gram of fuel, with a density of 10.2 g/cm^3) indicate that the specific leach rates vary from nuclide to nuclide. The data also indicate that leach rates are functions of the flushing rates for rates greater than one flush per year, while for flushing rates less than one flush per year the leach rates are essentially constant. The data on temperature effects currently available indicate that leach rates depend on temperature through an Arrhenius-type relationship. Finally leach rate data comparisons between solid material and crushed material indicate that leach rates are dependent on the actual surface area but not directly proportional to it since the internal surface area of the crushed waste is not as effective in leaching.

In the model, the highest specific leach rate was used for all nuclides and the entire fuel rod was assumed to leach at that rate. A total of 1000 metric tons of heavy metal was assumed to be crushed to 0.1 cm spheres. The leach function calculated by the model, as shown in Figure 3, was used as input to the mass transport model.

Hydrologic Model

Flow in the upper aquifer was simulated using a two-dimensional finite difference model (Variable Thickness Transient or VTT model) (Kipp et al. 1972) based on the Boussinesq equation. The model boundaries appear in

FIGURE 3. Release Rate Curve for Spent Fuel

Figure 1. The finite difference grid consists of 54 columns and 73 rows of nodes with a spacing of 7500 ft (2286 m). A submodel of the area surrounding the repository was also analyzed with a finer grid spacing. An auxilliary program was used to calculate the ground water velocities and travel times in flow tubes originating at the repository. Five such flow tubes are shown in Figure 4. The fifth flow tube constitutes the fastest pathway from the repository to the "S" River. Its length is 179,438 ft (34 miles or 54 km). The average ground water velocity is 4.16 ft/yr (4×10^{-6} cm/sec) and the average travel time is 43,000 years.

Mass Transport Model

The Multicomponent Mass Transport (MMT) (Ahlstrom et al. 1977) model was used to calculate the transport of radionuclides from the repository. This program accounts for simple radioactive decay, linear chain decay, branch chain decay, hydrodynamic dispersion and adsorption.

FIGURE 4. Ground Water Flow Tubes and Streamlines from the Breached Repository to the S River

A one-dimensional version of MMT was used along each flow tube to determine the contaminant concentrations at the discharge point as a function of time or the contaminant concentration distributions along the flow path.

The source term is shown in Table 1. The only isotopes considered are those which remain in significant amount after 10,000 years since the travel time of water to the "S" River exceeds 40,000 years. The isotopes which are not involved in chain decay can be followed one by one in the flow system.

TABLE 1. Initial Inventory

	ISOTOPE	HALF LIFE (YEARS)	K_d (mL/g)	UNCONTROLLED WATER RADIATION STANDARD (μ Ci/mL)	10 YEAR INVENTORY (CURIES/MTHM)
ACTIVATION AND FISSION PRODUCTS	NI-59	8.0E+4	50.	2.0E-4	3.03
	SE-79	6.5E+4	2.	3.0E-4	3.5E-1
	RB-87	4.7E+10	1.0	1.0E-4	1.7E-5
	MO-93	3.0E+3	15.	2.0E-4	7.3E-3
	NB-94	2.0E+4	80.	1.0E-4	1.0E-4
	ZR-93	9.5E+5	80.	8.0E-4	1.79
	TC-99	2.13E+6	1.9	3.0E-4	13.007
	PD-107	6.5E+6	38.	9.0E-5	9.9E-2
	SN-126	1.0E+5	80.	2.0E-5	.48
	I-129	1.59E+7	.5	6.0E-8	.033
	CS-135	2.3E+6	1.0	1.0E-4	.27
ACTINIDE CHAIN 1	PU-240	6540.	80.	5E-6	452.5
	U-236	2.3E+7	2.9	3E-5	.22
	TH-232	1.4E+10	80.	2E-6	1.1E-10
ACTINIDE CHAIN 2	CM-245	8.5E+3	1120.	4E-6	.18
	PU-241	15.	80.	2E-4	6.9E+4
	Am-241	433.	1130.	4E-6	1600.
	NP-237	2.14E6	23.	3E-6	.32
	U-233	1.58E5	2.9	3E-5	3.8E-5
	TH-229	7340.	80.	7E-6	2.8E-8
ACTINIDE CHAIN 3	CM-246	4760.	1120.	4E-6	3.5E-2
	PU-242	3.87E5	80.	5E-6	1.6
	U-238	4.47E9	2.9	4E-4	.32
	U-234	2.44E5	2.9	3E-5	.804
	TH-230	7.7E4	80.	2E-6	4.1E-6
	RA-226	1600.	.7	3E-8	7.4E-9
ACTINIDE CHAIN 4	AM-243	7.37E3	1130.	4E-6	14.7
	PU-239	2.439E4	80.	5E-6	290.
	U-235	7.04E8	2.9	3E-5	.016
	PA-231	3.25E4	100.	9E-7	6.35E-6

These include activation and fission products. The actinides, however, present linear and branch chain decay. The principal isotopes in these chains which were used for this exercise are shown in Tables 2 and 3.

RESULTS

Base Case

Figures 5, 6 and 7 show the time variations in the concentrations of Cs-135, I-129, and Tc-99 as they reach the river. These isotopes remain below MPC levels for uncontrolled water use. Other isotopes decay to negligible amounts before reaching the discharge point. As an example, the concentration

TABLE 2. Modeled Actinide Chains 2, 3 and 4

Cm-245	8.5E+3 Y
↓	
Pu-241	15.0 Y
↓	
Am-241	433.0 Y
↓	
Np-237	2.14E+6 Y
↓	
U-233	1.58E+5 Y
↓	
Th-229	7340.0 Y

ACTINIDE CHAIN 2

Cm-246	4760.0 Y	Am-242 M	152 Y
↓		↓	
Pu-242	3.87E+5 Y	Cm-242	0.45 Y
↓		↓	
U-238	4.47E+9 Y	Pu-238	87.8 Y
↘		↘	
U-234	2.44E+5 Y		
↓			
Th-230	7.7E+4 Y		
↓			
Ra-226	1600.0 Y		

ACTINIDE CHAIN 3

Cm-247	1.54E+7 Y		
↓			
Am-243	7.37E+3 Y	Cm-243	28 Y
↘		↘	
Pu-239	2.439E+4 Y		
↓			
U-235	7.04E+8 Y		
↓			
Pa-231	3.25E+4		

ACTINIDE CHAIN 4

of U-236, Th-230 and Ra-226 are shown in Figures 8, 9 and 10 versus the fractional distance to the discharge site.

Sensitivity Analysis

The purpose of this analysis is to examine how variations in the values of the input parameters influence the results. The parameters considered in the hydrologic model were the porosity and permeability of the upper aquifer, the boundary recharge rate and the fracture flow rate. In the mass transport model, the influence of retardation factor, dispersivity and ground-water velocity were studied. The sensitivity analysis for waste leach rates is currently being performed. Other parameters which can be accurately estimated or have little impact on the final results were considered to be deterministic. In this study each parameter was allowed to vary over a range of practical values while all other parameters were held constant at their base case value.

TABLE 3. Complete Actinide Chain No. 1

(UNDERLINED ISOTOPES
REPRESENT THE ONES MODELED)

Hydrologic Model Sensitivity Analysis

Porosity. Porosities equal to 1, 10 and 20 percent were used in the sensitivity analysis, although the base case value of 5 percent is thought to be representative of limestone and shale. As expected, there is a linear relationship between travel time and porosity since the ground water velocity is inversely proportional to the porosity (Figure 11).

KD= 1.00 ML/G HALF-LIFE=2.3000E+06 YEARS BETA= 35.0 G/ML

FIGURE 5. Concentration vs Time, Cs-135

KD= 0.50 ML/G HALF-LIFE=1.5900E+07 YEARS BETA= 35.0 G/ML

CONCENTRATION VS TIME

FIGURE 6. Concentration vs Time, I-129

KD= 1.90 ML/G HALF-LIFE=2.1300E+05 YEARS BETA= 35.0 G/ML

FIGURE 7. Concentration vs Time, Tc-99

KD= 2.90 ML/G HALF-LIFE=2.3000E+07 YEARS BETA= 35.0 G/ML
INVENTORY(CURIES) INITIAL=2.2000E+02.ON SOIL=3.2508E+02.IN THE WATER=3.2028E+00
CONCENTRATION VS DISTANCE AT 2.00E+06 YEARS

FIGURE 8. Concentration vs Distance, U-236

KD= 80.00 ML/G HALF-LIFE=7.7000E+04 YEARS BETA= 35.0 G/ML
INVENTORY(CURIES) INITIAL=4.1000E-03, ON SOIL=3.1703E+02, IN THE WATER=1.1323E+01
CONCENTRATION VS DISTANCE AT 2.00E+06 YEARS

FIGURE 9. Concentration vs Distance, Th-230

KD= 0.70 ML/G HALF-LIFE=1.6000E+03 YEARS BETA= 35.0 G/ML
INVENTORY(CURIES) INITIAL=7.4000E-06, ON SOIL=3.0368E+02, IN THE WATER=1.2395E+01
CONCENTRATION VS DISTANCE AT 2.00E+06 YEARS

FIGURE 10. Concentration vs Distance, Ra-226

FIGURE 11. Porosity vs Water Travel Time

Permeability. The only measured values of hydraulic conductivity for the upper aquifer were 0.3 ft/yr (2.9×10^{-7} cm/sec) and 102 ft/yr (9.9×10^{-5} cm/sec). The base case value was set at 100 ft/yr (9.7×10^{-5} cm/sec) and values of 0.3, 10, and 1000 ft/yr (2.9×10^{-7} , 9.7×10^{-6} and 9.7×10^{-4} cm/sec) were considered in the sensitivity analysis. Increasing the hydraulic conductivity increases the velocity in the upper aquifer as well as the rate of flow through the fracture. Thus, these two effects were considered simultaneously. Results are reported in Figure 12. It is interesting to note that the relationship between travel times and hydraulic conductivity is approximately linear on a log-log scale.

Boundary Recharge. In the base case, the northwest boundary of the model was held at a constant potential. The amount of recharge was calculated at each node, then increased by 50 percent and 100 percent for the sensitivity study. Figure 13 shows the corresponding changes in water velocities and travel times for the third flow tube. Increasing the recharge increases the velocity but also alters the potential distributions thereby reducing the flow path length to the discharge site. It is found that the travel time between the repository and the discharge site is reduced by a factor 3 when the recharge is doubled.

Fracture Flow Rate. The base case flow rate of 98.18 gpm (6.19 l/sec) through the entire length of the fracture was increased by 63 percent to 160 gpm (10 l/sec) which was found to be the maximum flow rate compatible with the permeabilities of the upper and lower aquifers and the head difference between these systems. The results obtained indicate that this increased flow rate reduces the water travel time by only 5 percent.

Mass Transport Model Sensitivity Analysis

The sensitivity analyses were performed for the three fission products (Cs-135, I-129, Tc-99) that reach the river in measurable quantities and for the four actinide chains.

Dispersivity. The value of dispersivity set for the base case at 10 feet (3 m) was varied in the range of 0.01 ft (0.3 cm) to 100 ft (30 m). Increasing the dispersivity in this interval was found to lower significantly the peak

FIGURE 12. Permeability vs Water Travel Time

FIGURE 13. Water Velocity and Travel Time vs Increase in Boundary Recharge

concentrations but to have little effect on peak arrival times. This phenomenon is illustrated in Figure 14 for I-129. In this case it was found that increasing the dispersivity by four orders of magnitude reduces the peak concentration by a factor 12.

The sensitivity of the peak concentration on dispersivity was analyzed for the three fission products Cs-135, I-129, and Tc-99 using their base case retardation factors ($K=36$, 18.5 , and 67.5 , respectively) and for I-129 using a range of retardation factors. Figure 15 shows the decrease in peak concentration of these isotopes as dispersion increases. The peak concentrations are scaled to the values obtained with a dispersivity of 0.01 ft (0.3 cm) which represents virtually zero dispersion. It is interesting to note that the effect of dispersion on peak concentration is very similar for isotopes having different half lives but identical retardation factors. Figure 15 also illustrates that dispersion and retardation factors have a similar influence on peak concentrations. For instance, the reductions in Iodine peak concentrations achieved with K equal to 18.5 and dispersivity equal to 100 ft (30 m) is the same as the reduction obtained with K equal to 67.5 and a dispersivity of 10 ft (3 m).

Retardation Factor. The retardation factor K is a function of the adsorption coefficient K_d and of the soil to solution ratio B , i.e., $K = 1 + BK_d$. Because B and K_d have a similar influence on K , the sensitivity analysis was made on K and thus illustrates the effect of both K_d and B parameters. The Figure 16 for the three fission products as a single straight line on a log-log scale. This indicates that the peak arrival time is a direct function of the retardation factor.

Any parameter affecting peak arrival time will also affect the peak concentrations because the total time available for decay will be altered. In addition, there is a complex interaction between dispersivity and retardation factors. All other input parameters being constant, there is a threshold retardation factor such that any increase in this retardation factor (even after correcting for the effect of decay) causes a reduction in peak concentration. Above this threshold increasing the retardation factor appears to

KD= 0.50 ml/g HALF-LIFE = 1.5900E+07 YEARS BETA= 35.0 g/ml FLOW TUBE 1
CONCENTRATION VS TIME

FIGURE 14. Concentration vs Time, ^{129}Tc

FIGURE 15. Decrease in Peak Concentration as Dispersion Increases

amplify the effect of hydrodynamic dispersion. This point is illustrated in Figure 17 where the peak concentration is plotted versus retardation factor for different values of dispersivity. The isotope is I-129 and the results are scaled to those corresponding to the case of no retardation ($K=1$). It is seen that a larger retardation factor results in lower peak concentration. Similarly, for a given retardation factor, a larger dispersivity also reduces peak concentration.

Water Velocity. The relationships between ground water velocity and peak arrival time are shown in Figure 18 for the isotopes Cs-135, I-129, and Tc-99. These relationships are plotted as parallel straight lines on a log-log scale, which reflects the fact that those isotopes have different K_d 's.

FIGURE 16. Time vs Retardation Factor

Variations in water velocity influence the peak concentration because they modify the time available for radioactive decay and because of dilution effects. The relationships between water velocity and peak concentrations are shown in Figure 19 for Cs-135, I-129, and Tc-99. In the case of Tc-99, which has a shorter half life than the other two fission products, the results show that increasing the velocity increases the peak concentration, i.e., the effect of radioactive decay is predominant. For isotopes such as I-129 and Cs-135, which have longer half lives, the results show the combined influences of dilution, dispersion, and radioactive decay. The effect of dilution is illustrated in Figure 20 for the case of I-129. If the flow tube width is adjusted to maintain a constant discharge to the river, dilution effects are eliminated. The result is an increase in peak concentration with increased velocity. Conversely, if dilution alone is considered there is an increase in peak concentration with decreasing water velocities. The addition of these two phenomena yields the curve labeled "total effect" on Figure 20.

FIGURE 17. Dispersion vs Retardation Factor

FIGURE 18. Relationship Between Ground Water Velocity and Peak Arrival Time

FIGURE 19. Relationship Between Water Velocity and Peak Concentration

FIGURE 20. Total Effect of Increase in Peak Concentration with Increased Velocity

CONCLUSIONS

A numerical analysis of the consequences of radionuclide release from a hypothetical nuclear waste repository located in bedded salt and of their subsequent transport by ground water has been performed. The study was based on available geologic and hydrologic information for the reference location. The initial run was based on a best estimate of geologic and hydrologic parameters for this reference location. Its results indicate that those isotopes which reach the natural discharge point remain below MPC levels for uncontrolled water use. The initial run was followed by a parametric study to evaluate the sensitivity of the model to the values of input parameters. This analysis shows that the model is sensitive to the assumed values of hydraulic conductivity, dispersivity and retardation factor, as expected. But this analysis also emphasizes that there exist complex interactions between such parameters which are less intuitively evident. The influence of waste leach rates is being investigated.

In conclusion, this exercise demonstrates that numerical modeling can assist in providing an evaluation of geologic formations as nuclear waste repositories and delineate the areas where further research is warranted.

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