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ONE-DIMENSIONAL RADIONUCLIDE TRANSPORT UNDER TIME-VARYING CONDITIONS*

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ABSTRACT

New analytical and numerical solutions are presented for one-dimensional radionuclide transport under time-varying fluid-flow conditions including radioactive decay. The analytical solution assumes that all radionuclides have identical retardation factors, and is limited to instantaneous releases. The numerical solution does not have these limitations, but is tested against the limiting case given for the analytical solution. Reasonable agreement between the two solutions was found. Examples are given for the transport of a three-member radionuclide chain transported over distances and flow rates comparable to those reported for Yucca Mountain, the proposed disposal site for high-level nuclear waste.

I. INTRODUCTION

The impact of assuming steady-state flow conditions on the simulation of radionuclide transport at high-level nuclear waste (HLW) disposal sites has not been established. For example, over the regulatory time frames of interest, typically thousands of years, one may expect changes in the fluid-flow rates through the repository due to changes in infiltration and recharge caused by intermittent rainfall or climatic changes. Such changes are difficult to predict, but that does not eliminate the need to assess how such changes may affect results from models that assume steady-state flow conditions.

To address part of this problem, the computer code NEFTRAN (NEtwork Flow and TRANsport)[1] has been modified and an analytical solution has been developed to test this modification for time-varying flow conditions on

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the transport of a decaying radionuclide chain.[1] Few analytical solutions are available for time-varying conditions,[2,3,4,5,6] and no analytical solutions which consider radioactive decay with time-varying flow rates have been found in the literature. Furthermore, for time-varying conditions, even without radioactive decay, no analytical analysis has been found on the cumulative release of radionuclide past a fixed point in space. Such an analysis is important because the performance measure in the containment requirements of the Environmental Protection Agency's standard for the disposal of high-level, spent fuel, and transuranic wastes is given in terms of cumulative radionuclide releases to the accessible environment over 10,000 years.[7] The analysis in this paper extends available analytical solutions to include the instantaneous release of decaying radionuclides from a repository of arbitrary length under time-varying flow conditions, assuming that the radionuclides have identical retardation factors. This solution has been extended to calculate the cumulative release of radionuclides past a given fixed point in space. The numerical solution is more general and does not assume identical retardation factors, and the source rate is not limited to instantaneous releases from the repository.

II. ANALYTICAL SOLUTION

For time-varying one-dimensional convective and dispersive transport of a radionuclide chain through an adsorbing porous medium, the governing equation for a chain of m radionuclides is,[8,9]

$$R_i \frac{\partial C_i}{\partial t} + U(t) \frac{\partial C_i}{\partial x} = D(t) \frac{\partial^2 C_i}{\partial x^2} - R_i \lambda_i C_i + R_{i-1} \lambda_{i-1} C_{i-1} \tag{1}$$

where $-\infty < x < \infty$,

$i = 1, \ldots, m$,

$\lambda_o = 0$,

t is time,

C_i is the molar concentration in solution of the i-th radionuclide,

R_i is the retardation factor of the i-th radionuclide,

λ_i is the radioactive decay rate constant of the i-th radionuclide,

U(t) is the fluid-flow velocity, and

D(t) is the dispersion coefficient.

To model time-varying flow, U(t) and D(t) are assumed to be time dependent. The retardation factor is given by

$$R_i = 1 + \frac{\rho_s K_{di}}{\phi} \tag{2}$$

where ϕ is the porosity, ρ_s is the bulk density and K_{di} is the adsorption distribution coefficient for the i-th radionuclide.

The solution for a repository of length h releasing radionuclides of a concentration $C_{i,s}$, at t=0, in the region $-h \le x \le 0$, for $R_i = R$, $i=1, \ldots, m$, may be constructed from the Green's function, and is given by,[10]

$$C_i(x,t) = \frac{1}{2}\left\{ \operatorname{erf}\left[\frac{x+h-\bar{U}t}{\sqrt{4\bar{D}t}}\right] - \operatorname{erf}\left[\frac{x-\bar{U}t}{\sqrt{4\bar{D}t}}\right]\right\}$$

$$\cdot \sum_{j=1}^{i} \alpha_j b_i^{(j)} \exp(-\lambda_j t) \qquad (3)$$

where the time-averaged species velocity and retarded dispersion coefficent are given respectively by

$$\bar{U}_i = \frac{\int_0^t U(\tau)\, d\tau}{Rt} \qquad (4)$$

and

$$\bar{D}_i = \frac{\int_0^t D(\tau)\, d\tau}{Rt} \qquad (5)$$

the i-th element of the j-th eigenvector is given by

$$b_i^{(j)} = \begin{cases} 0 & i < j \\ 1 & i = j \\ \prod_{k=j}^{i-1} \dfrac{\lambda_k}{\lambda_{k+1} - \lambda_j} & i > j \end{cases} \qquad (6)$$

and

$$\alpha_1 = C_{1,s} \qquad (7)$$

$$\alpha_i = C_{i,s} - \sum_{j=1}^{i-1} \alpha_j b_i^{(j)} \qquad i > 1 \qquad (8)$$

As discussed previously, a performance measure for HLW disposal systems is the cumulative radionuclide mass reaching the accessible environment over a specified length of time. This quantity for one-dimensional transport of the i-th radionuclide past the point x = L is given by the cross-sectional area for flow times $f_i(t)$, where $f_i(t)$ is given by

$$f_i(t) = \int_0^t \frac{U(\tau)C_i(L,\tau)}{R_i} - \frac{D(\tau)}{R_i}\frac{\partial C_i(L,\tau)}{\partial x} \, d\tau \quad (9)$$

L is taken as the location of the accessible environment, and $f_i(t)$ is the cumulative sum of the convective and dispersive mass fluxes of radionuclide i. Since $U(\tau)$ and $D(\tau)$ are arbitrary functions of time, the integral in Equation (9) can not be evaluated until these functions are specified. Furthermore, numerical integration may be required since $C_i(L,\tau)$, $U(\tau)$, and $D(\tau)$ may be given in terms of complicated functions that are not explicitly integrable.

III. NUMERICAL SOLUTION

To extend the analysis for the more realistic conditions where the retardation factors for each radionuclide are not identical, a numerical solution may be obtained by using a modified version of the NEFTRAN code. NEFTRAN was developed originally to simulate transport of radionuclide chains in a steady-state flow field represented by a series of one-dimensional legs. Each leg may have different physical and chemical properties. Transport is modeled using the Distributed Velocity Method (DVM),[11] which consists of dividing each leg into equal-sized grid blocks. At the beginning of each time step, the atoms in each grid block are uniformly distributed in space. For each radionculide, these atoms are partitioned equally into several packets. Each packet is then assigned a different migration velocity. These migration velocities have a Gaussian distribution with the mean equal to the species velocity, and the standard deviation determined from the dispersion coefficient. At the end of the time step, the distance traveled by a packet is equal to the product of the time step and the packet migration velocity. This distance plus the initial starting point determines which grid blocks the packets overlaps. At the end of the time step, all atoms that reach a grid block are summed to form the initial number of atoms in the grid block for the next time step.

Radioactive decay and production over the time step are included in the model by determining the fraction of atoms that survive decay and the fraction that produce daughter products over the time step. Transport of atoms that change identity over a time step is accomplished using species-averaged velocities. In addition to the DVM transport model, NEFTRAN contains a steady-state network flow-solver and several radionuclide source term models.

NEFTRAN has been modified to simulate unsteady flow by approximating the change in fluid-flow velocity with time as discrete sequences of steady-state flows, and uses the DVM to transport radionuclides over time intervals of constant flow. Comparisons of this new numerical approach with an analytical solution are given in the next section.

IV. APPLICATIONS

In this section, two examples are presented. The input data were taken from preliminary studies for the proposed site at Yucca Mountain, the only site currently under consideration for a HLW repository in the U. S. However, these examples are only adequate to compare the analytical solution with the modified version of NEFTRAN, and not to assess the performance of this particular site. The total simulation time for both of these examples was 100,000 years. This is inconsistent with the EPA standard which is based on 10,000 years, but was necessary to implement the comparison. The first example is for uniform retardation factors with a doubling of the fluid-flow velocity at an arbitrarily determined time of 50,000 years into the simulation. In this example, the analytical and numerical solutions are compared. In the second example, the retardation factors depend on the radionuclide, and thus only the numerical solution is presented.

The numerical and analytical solutions for the discharge rates are given in Figure 1 for the transport of the three member chain of Am-243, Pu-239 and U-235, with half-lives of 7593 years, 24,400 years, and 7.1×10^{8} years, respectively, and initial inventories of 9.1×10^{5} Ci, 1.96×10^{7} Ci, and 1.12×10^{3} Ci, respectively. These inventories correspond to the amount of HLW that may be placed in the proposed repository at Yucca Mountain.[12] The migration-path length was assumed to be 200 meters, the length from the proposed repository horizon at Yucca Mountain to the water table.[13] A constant fluid-flow velocity of 3.0 cm/year was assumed for the flow rate.[13] The dispersion coefficient was assumed to be proportional to the fluid-flow velocity with a dispersivity of 20 meters. At 50,000 years the fluid-flow velocity was doubled. A retardation factor of 19 was assumed for all the radionuclides. This was the lowest value reported[12] for these radionuclides for Yucca Mountain tuff. Although all of these values have been reported in the literature for Yucca Mountain, extensive laboratory and field experiments have not been conducted, and these are only preliminary values.

As shown in Figure 1, the overall agreement between the analytical and the numerical solutions is reasonable. The discharge rates estimated with NEFTRAN are generally larger than those for the analytical solution. However, the shape of the discharge curves are consistent. For all three radionuclides, NEFTRAN also predicts larger integrated discharges (between 30% and 50%) to the accessible environment at 100,000 years compared to the analytical solution. This discrepancy can not be explained at this time, but will be addressed before the modified version of NEFTRAN is released.

In the second example, all parameter values were equal to those in the first example, except for the retardation factors for Am-243 and Pu-239 which were changed to 1800 and 640, respectively, as reported for Yucca Mountain tuff.[12] For this case, only a numerical solution is available, and is shown in Figure 2. Comparing Figures 1 and 2, the increase in retardation factors greatly reduces the discharge rates.

V. SUMMARY AND CONCLUSIONS

An analytical solution has been obtained for one-dimensional radionuclide transport under time-varying fluid-flow velocities and dispersion coefficients, including radioactive decay. This solution is limited to instantaneous radionuclide releases and identical retardation factors for all radionuclides. Numerical solutions are required to account for retardation factors that are a function of the species and for time-varying sources. A modified version of the NEFTRAN code was tested by using the analytical solution in this work for cases when the retardation factors are uniform for an instantaneous source. For the example in this work, it was shown that in comparison to the analytical solution, NEFTRAN provides reasonable agreement.

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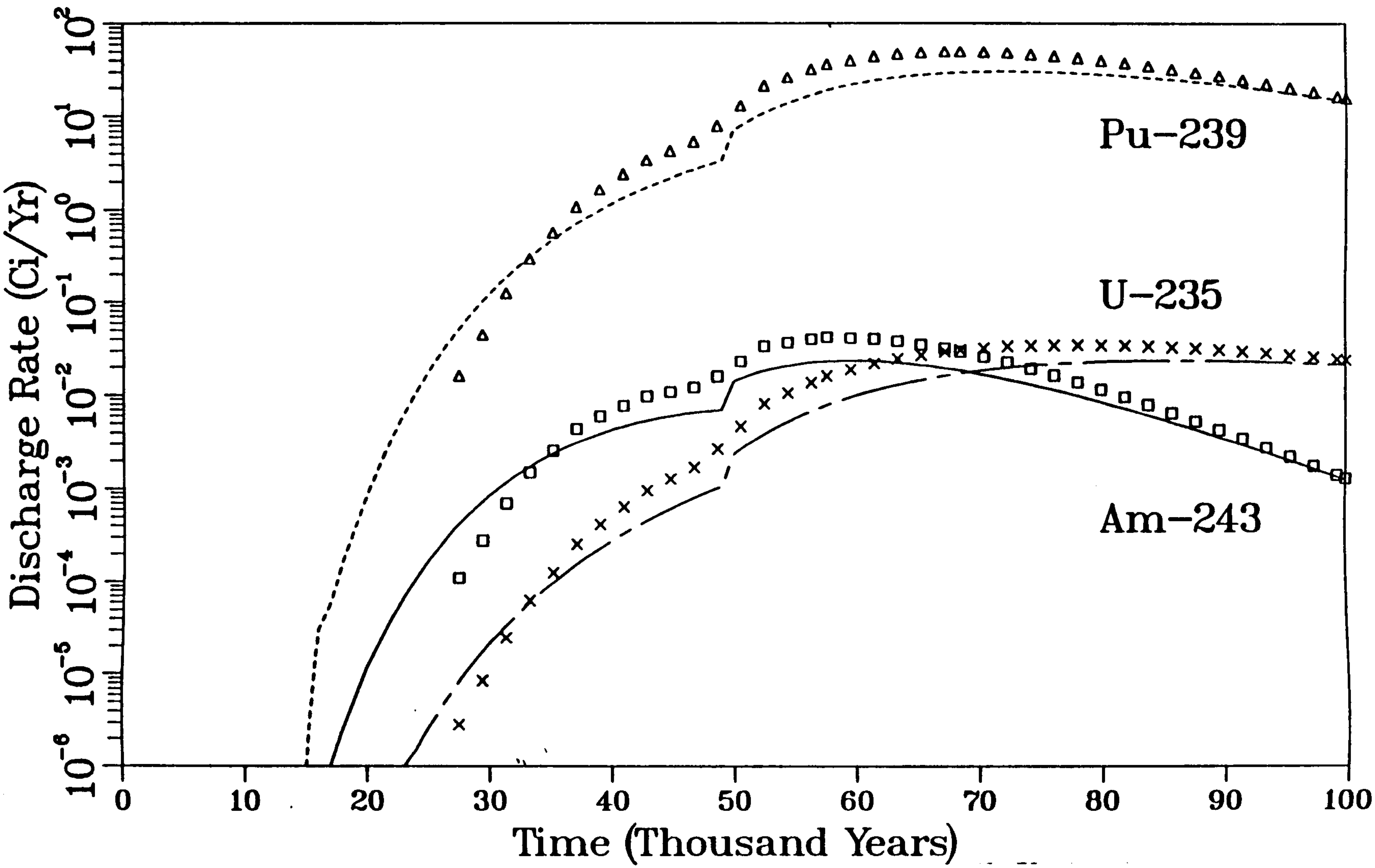

Figure 1. Comparison of analytical and numerical solutions given by lines and points, respectively for the first example problem of a step change in the fluid-flow velocity at 50,000 years, with uniform retardation factors.

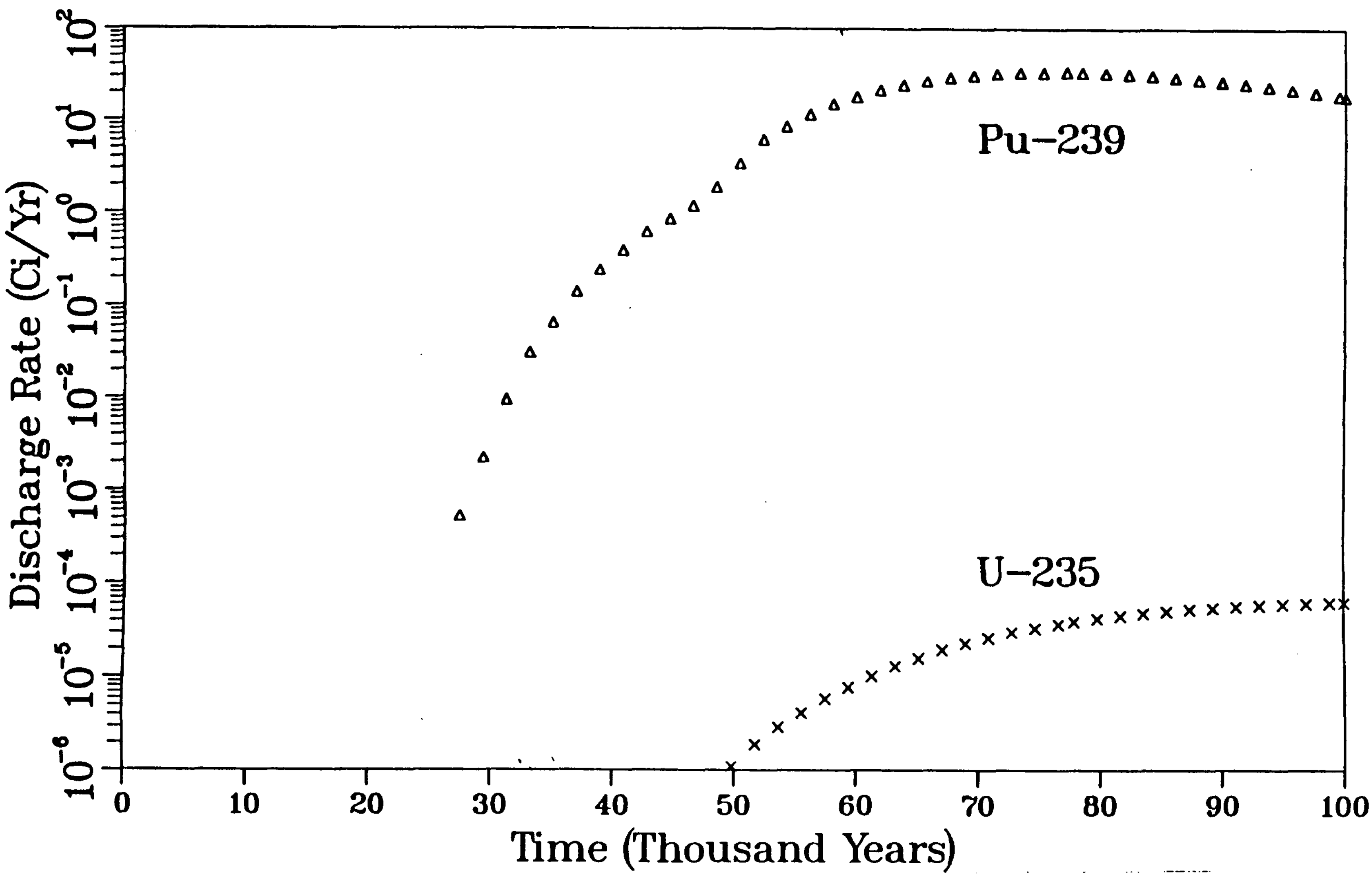

Figure 2. Numerical solution for the second example problem of a step change in the fluid-flow velocity at 50,000 years, with nonuniform retardation factors. No significant amount of Am-243 was discharged.