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Authors

Sadeghi, M.M.

Lee, W.W.-L.

Pigford, T.H.

et al.

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Release Rates of Radionuclides into Dripping Ground Water

M. M. Sadeghi, W. W.-L. Lee, T. H. Pigford, and P. L. Chambre

Earth Sciences Division
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

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RELEASE RATES OF RADIONUCLIDES INTO DRIPPING GROUND WATER**M. M. Sadeghi, W. W.-L. Lee, T. H. Pigford, P. L. Chambré****Department of Nuclear Engineering, University of California****and****Lawrence Berkeley Laboratory
University of California****Berkeley, CA 94720****Introduction**

Packages of high-level waste are to be emplaced in unsaturated tuff at the proposed Yucca Mountain repository. Each cylindrical waste package is separated from surrounding rock by a 2-cm air gap. A possible mechanism for release of radionuclides is the dripping of ground water onto waste packages. Drips are assumed to penetrate cracks in failed container and to dissolve radionuclides as the partly failed container fills and overflows.¹ For this wet-drip scenario, with assumed constant drip rate, we have developed analytical solutions for the time-dependent release rates of radionuclides to the surrounding rock.² Release rates have been calculated for key radionuclides³ using parameter values from references 4 and 5.

Solubility-Limited Releases

For a low-solubility species, the release rate after container overflow is the product of the volumetric drip rate, the elemental solubility, and the time-dependent isotope fraction of that species. Division by the 1000-year inventory gives the NRC-defined fractional release rate. Typical results for plutonium isotopes released from spent fuel are shown in Figure 1. For the parameters indicated on the figure, it takes about 8600 years to fill the container. As Pu-240 decays, the longer-lived isotopes dissolve to a greater fraction of the elemental solubility. The release rate of Pu-239 increases with time until attenuated by decay, after about 60,000 years.

Releases of Readily Soluble Species

Fission-products such as cesium and iodine in fuel-cladding gaps, plenum, and grain boundaries are assumed to dissolve quickly when contacted by water filling the container. Assuming ten percent of the total inventory of these species is available for instant dissolution, and assuming thorough mixing of the liquid within the container, we obtain the time-dependent fractional release rates of these species shown in Figure 2.

Alteration-Controlled Releases

Oxygen in ground water and hydrogen peroxide from α -radiolysis are assumed to react slowly with the spent-fuel matrix, resulting in an altered uranium structure. Species in the spent-fuel matrix are assumed to be released congruently with the alteration rate, if not limited by their own solubility. For an assumed constant fractional alteration rate of 0.001/yr, a linear increase in the fuel surface undergoing alteration as the container fills, and no solubility limit for technetium, we obtain the time-dependent fractional release rate for Tc-99 shown in Figure 3.

Other Results

Our analyses have also included release rates of other key radionuclides and decay daughters of Pu-238 and U-234, for both spent fuel and borosilicate-glass defense waste.²

Conclusions

The calculated results are conservative to the extent that barriers from fuel cladding have not been considered. Also, repository-average release rates can be lower if the expected statistical distribution of container failures is included.

For the parameters adopted here, release rates of low-solubility species are favorably low. Release rates of readily-soluble species are higher but may not be limiting for repository-average release rates. Spent-fuel alteration may result in the most limiting release rates. Better experimental data are needed to define the long-term alteration rates for spent fuel and borosilicate glass.

Possible liquid diffusive pathways from the waste package into surrounding rock could augment the releases calculated from the wet-drip scenario.⁶ Results for such a wet-continuous environment are presented in a separate paper.⁷

References

1. W. J. O'CONNELL, D. A. LAPP, and R. M. THATCHER, "Waste Package Performance Assessment for the Yucca Mountain Project," Waste Management 89, Tucson (1989)
2. M. M. SADEGHI, T. H. PIGFORD, P. L. CHAMBRÉ and W. W.-L. LEE, "Equations for Predicting Release Rates for Waste Packages in Unsaturated Tuff," LBL Report, Lawrence Berkeley Lab. (1989).
3. M. M. SADEGHI, T. H. PIGFORD, P. L. CHAMBRÉ and W. W.-L. LEE, "Prediction of Release Rates for a Waste Repository at Yucca Mountain," LBL Report, Lawrence Berkeley Lab. (1989).
4. C. N. WILSON, and C. J. BRUTON, "Studies on Spent Fuel Dissolution Behavior Under Yucca Mountain Repository Conditions," PNL-SA-16832 (1989).
5. "Site Characterization Plan," Yucca Mountain Site, Nevada Research and Development Area, Nevada, DOE/RW-0199, U.S. Department of Energy (1988).
6. T. H. PIGFORD, and P. L. CHAMBRÉ, "Radionuclide Transport in Geologic Repositories: A Review," in M. J. Apted and R. E. Westerman (eds.), *Scientific Basis for Nuclear Waste Management XI*, 125, Pittsburgh, Materials Research Society (1987).
7. M. M. SADEGHI, W. W.-L. LEE, T. H. PIGFORD, and P. L. CHAMBRÉ, "Diffusional Release of Radionuclides into saturated and Unsaturated Tuff," LBL-28428, Paper submitted for the 1990 American Nuclear Society Annual Meeting.

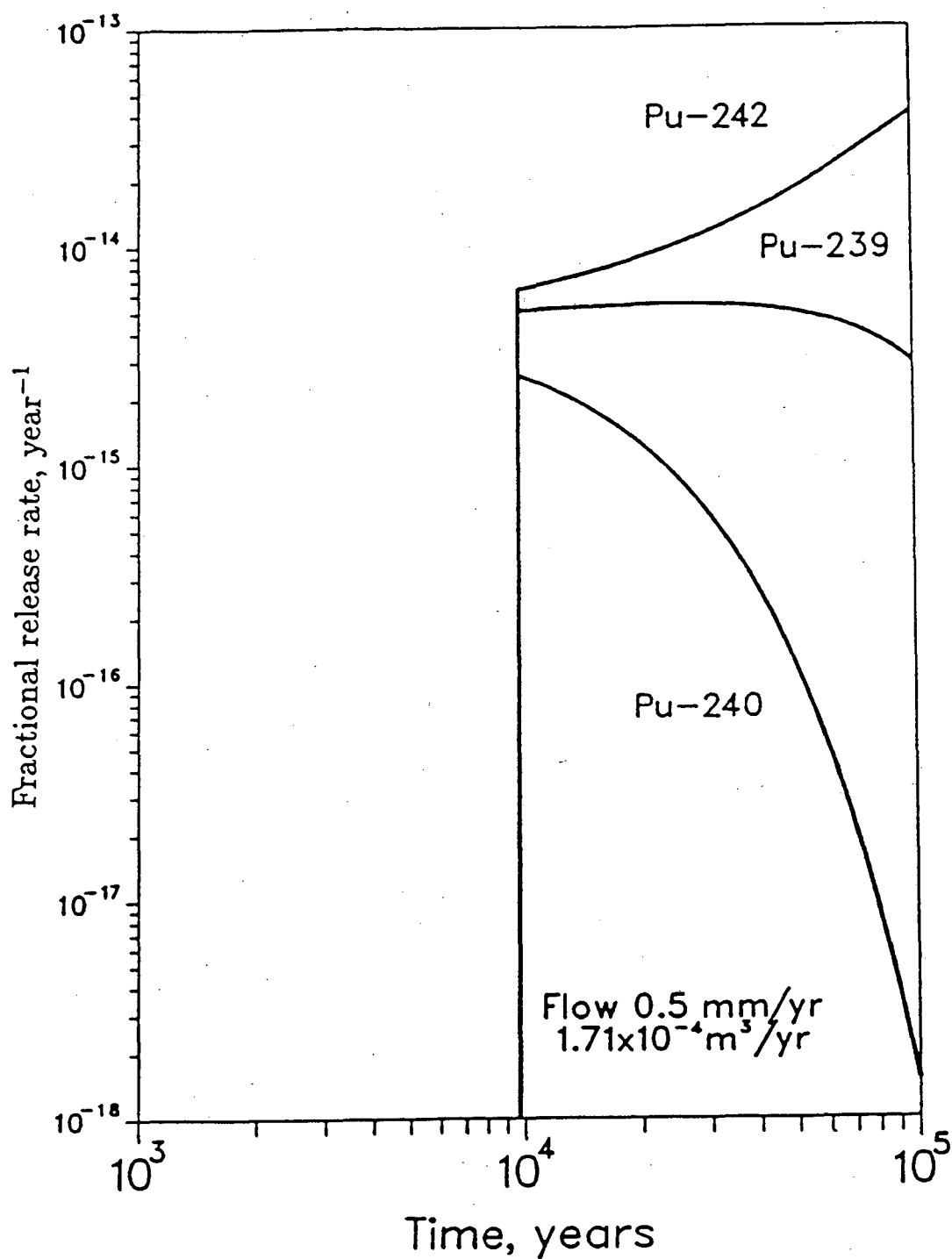


Fig. 1. Fractional release rate of plutonium isotopes from spent fuel, wet-drip scenario (container failure at 1000 years, Pu solubility= 2.9×10^{-7} g/m³, container void volume=1.5 m³)

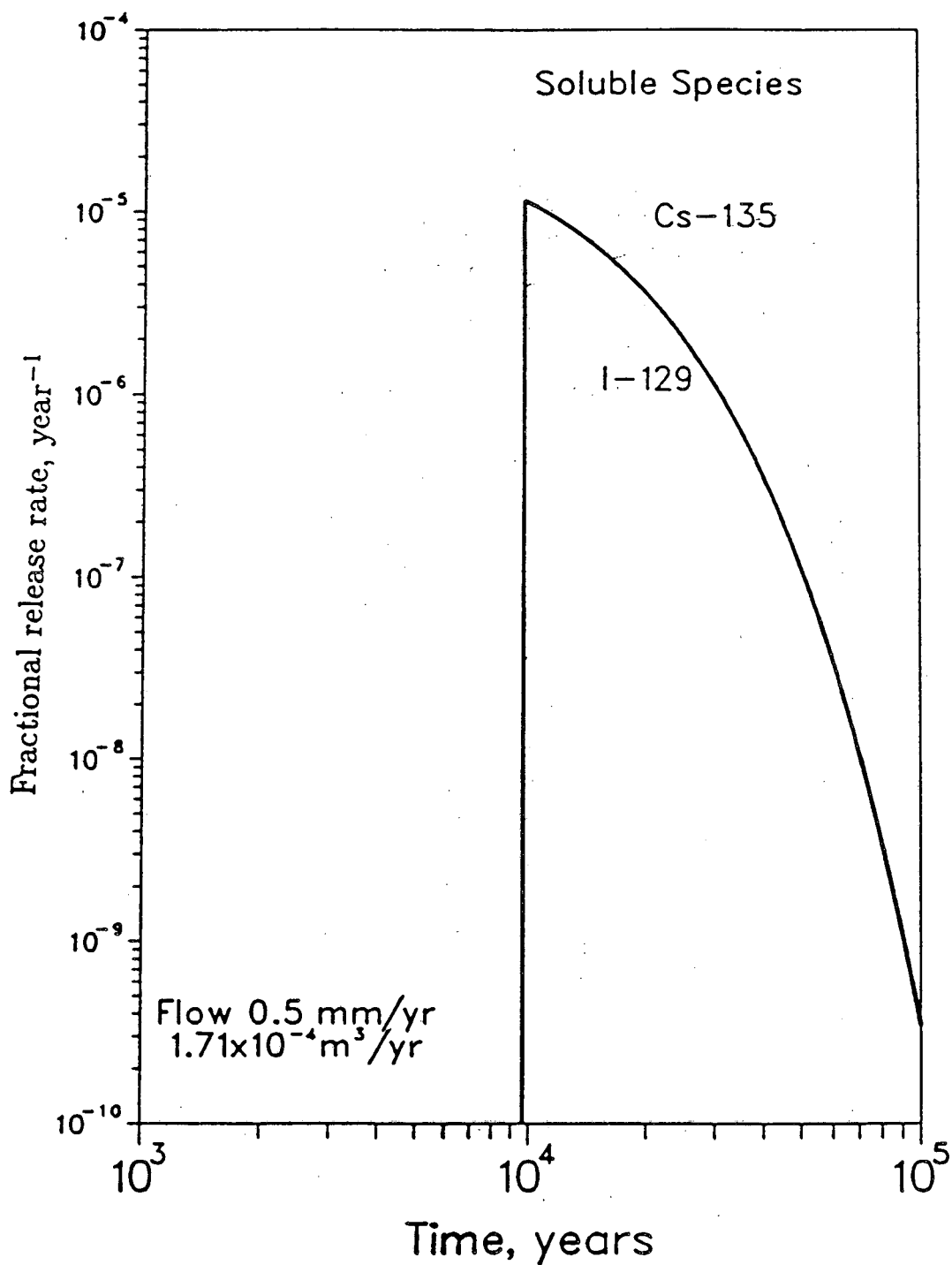


Fig. 2. Fractional release rate of readily soluble species from spent fuel, wet-drip scenario (container failure at 1000 years, 10 percent of Cs, I inventory is assumed to be readily soluble)

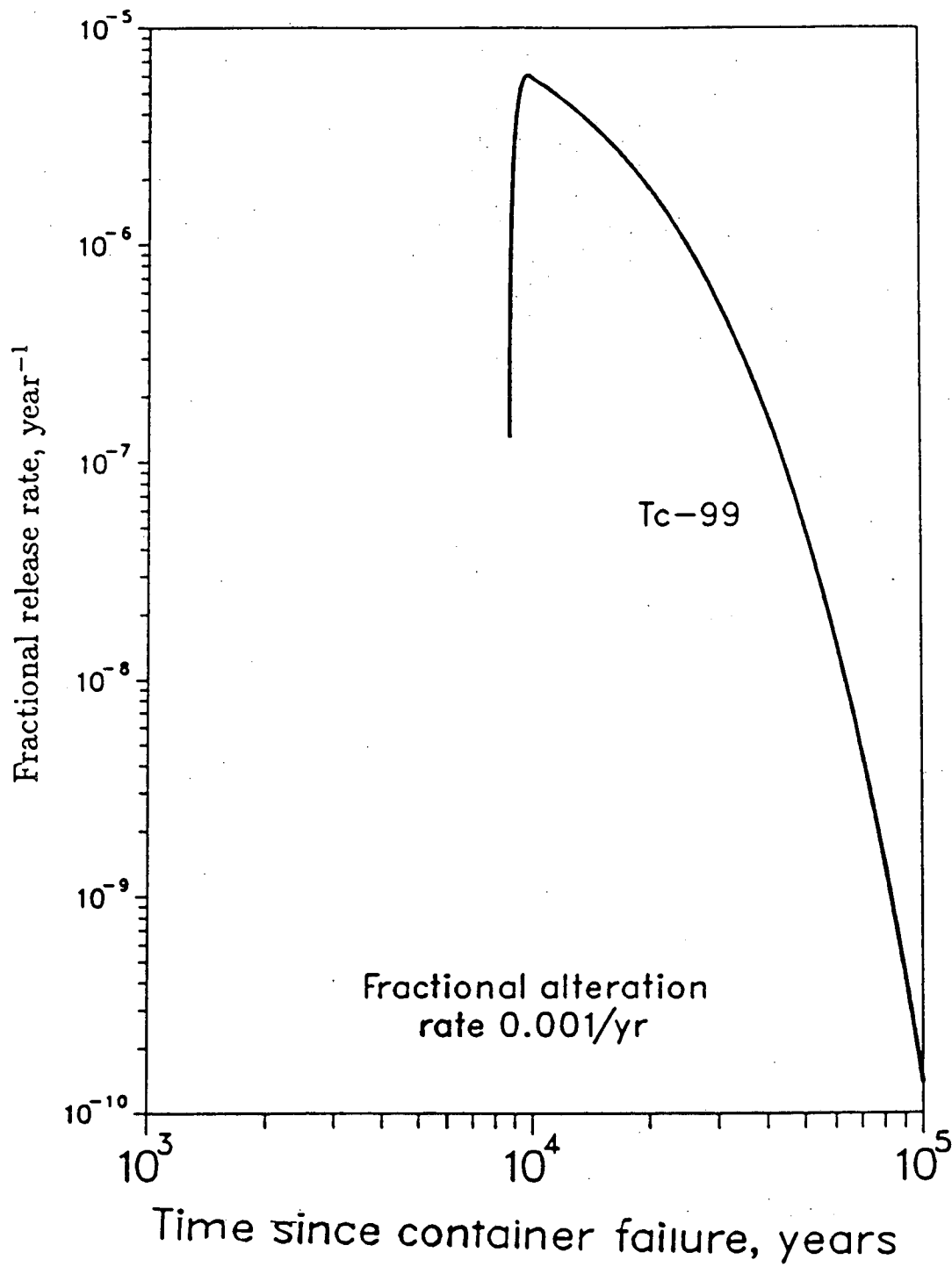


Fig. 3. Alteration-controlled fractional release rate of Tc-99 from spent fuel

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INFORMATION RESOURCES DEPARTMENT
BERKELEY, CALIFORNIA 94720