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Invited paper presented at the ASCE Conference, Energy '84, Pasadena, CA, August 19-22, 1984

ASSESSING THE PERFORMANCE OF GEOLOGIC REPOSITORIES FOR NUCLEAR WASTE

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August 1984

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

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UCB-NE-4051
LBL-18270

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This work was sponsored in part by the U. S. Department of Energy under Contract Number DE-AC03-76SF00098.

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INTRODUCTION

A recent study by the National Research Council (Pigford et al., 1983a) summarized the important features that affect the long-term performance of geologic repositories, and it illustrated a conceptual approach for predicting repository performance. The purpose is to predict where, when, and how much radionuclides from the waste reach the environment. Here we summarize predictions of the rate of release of radionuclides from waste packages, their rates of dissolution in groundwater, their hydrogeologic transport to the environment, and their ultimate uptake by people. Both the details of performance assessment and conclusions on performance are affected by what performance criteria are adopted.

DISSOLUTION RATES

Typical waste packages contain UO_2 fuel rods discharged from light-water reactors or borosilicate glass waste from the defense and commercial fuel cycles. The Nuclear Regulatory Commission (NRC) requires that these waste solids be protected from groundwater by a corrosion-resistant envelope that must last for 300 to 1,000 years, until most of the heat generation has decayed. It is difficult to extrapolate laboratory leach data to predict the rate of dissolution of the remaining radionuclides for the next few thousand years or more. Alternatively, the rate of mass transfer of individual radioelements into surrounding groundwater has been conservatively predicted by assuming solubility of each constituent at the waste surface and by solving the equations for diffusive-convective mass transfer in the surrounding flow field (Chambré et al., 1982a,b,c,1983). The solubilities of most of the important radionuclides are low enough that the mass-transfer rates are much lower than observed in laboratory leach experiments wherein the mass-transfer resistance of the flow field is not present. Calculated dissolution rates for specific radionuclides, normalized to the inventory of each radionuclide in the waste, are shown in Table 1 for a typical borosilicate glass waste surrounded by porous rock.

Table 1. Calculated Fractional Rates for Borosilicate Glass Waste in a Repository

Waste cylinder: radius = 0.152 m, length = 2.46 m, fission-product and actinide oxides from 460 kg of uranium in fuel, groundwater pore velocity = 1 m/yr.

Constituent	Concentration, g/cm^3	Solubility[a], g/cm^3	Predicted Fractional Dissolution rate, yr^{-1}
SiO_2	1.6	5×10^{-5}	1.1×10^{-6}
Tc	1.92×10^{-3}	1×10^{-9}	2×10^{-8}
U	1.22×10^{-2}	1×10^{-9}	4×10^{-9}
Np	1.92×10^{-3}	1×10^{-9}	2×10^{-8}
Pu	1.15×10^{-4}	1×10^{-9}	4×10^{-7}
Am	3.56×10^{-4}	1×10^{-10}	1×10^{-8}
Se	1.40×10^{-4}	1×10^{-9}	3×10^{-7}
Sn	9.40×10^{-5}	1×10^{-9}	5×10^{-7}

a/ From data summarized by Krauskopf (Pigford, 1983a)

Similar approaches to predicting waste dissolution rates have been adopted by Sweden's KBS-3 design study (SKBF, 1983) of geologic repositories and by the basalt waste isolation project in this country. In the Swedish design the million-year life of a protective copper canister is predicted by a similar calculation of the diffusion of sulfide oxidant from groundwater to the corroding surface, and mass-transfer calculations predict later radionuclide releases. More recent mass-transfer studies include reaction-rate data (Zavoshy et al., 1984) to provide a means for calculating concentrations at the waste surface and for estimating more realistic dissolution rates in a repository.

The NRC regulations for geologic disposal require that packages of high-level waste release individual radionuclides to groundwater at a rate no greater than 10^{-5}/year times the 1000-year inventory of that radionuclide, or at a rate no greater than 10^{-8}/yr times the total curie inventory at 1000 years. The calculations in Table 1 indicate that this may be easily met for the low-solubility radionuclides

An invited paper presented at "Energy '84", ASCE Conference, Pasadena, CA, August 19-22, 1984.

present in relatively high concentration in the waste solid, but it may be difficult to meet for more soluble species. This will be particularly a problem for unreprocessed spent fuel, because of iodine-129, carbon-14, and other likely soluble species in the UO_2 and because of carbon-14 and other activation products in the Zircaloy cladding, unless the Zircaloy presents a long-lived barrier that prevents or reduces radionuclide release. Locally oxidizing environments, that may occur from an oxygen environment or from radiolysis, can increase solubilities and release rates. Although sorbing backfill around the waste can delay the release of radionuclides to groundwater, backfill has little affect on the longer-term steady state release of long-lived radionuclides in wet-rock repositories. However, exceeding the NRC release-rate requirement does not necessarily contribute to a significant hazard from calculated environmental releases (Pigford et al., 1983a).

HYDROGEOLOGIC TRANSPORT TO THE ENVIRONMENT

After determining the radionuclide source terms, the hydrological flow field from the repository to the environment, and the sorptive properties of the geologic medium, the hydrogeologic transport of dissolved radionuclides can be calculated from the equations of advective transport. Whereas hydrogrologic transport is a likely long-term process for wet-rock repositories, such calculations for a salt repository must unrealistically assume intrusion of flowing water that dissolves the salt. Useful scoping estimates for wet-rock repositories assume one-dimensional advective transport along flow streamlines (Harada et al., 1980), with spatially independent coefficients of hydrodynamic dispersion, to predict the time-dependent concentrations of individual radionuclides reaching the environment.

It is difficult to estimate the distribution of radiation doses to future people who may be exposed to this contaminated groundwater, but a conservative approach is to calculate the dose to a maximally exposed individual, a person whose entire lifetime intake of potable water and food is affected by the contaminated water. Limiting the calculated dose to future maximally exposed individuals is the criterion adopted for geologic repository designs in other countries, e.g., Canada, Sweden. Using sorption data and concentration-dose conversion factors (Pigford et al., 1983a), we assume a repository loaded with reprocessing waste and calculate the average annual radiation doses from radionuclides released to the environment, normalized to the maximum dose rate from carbon-14, as shown in Figure 1. A water travel time of 1000 years is conservatively assumed; in typical wet-rock repository sites the mean time for contaminated groundwater to reach the environment is of the order of 10^4 to over 10^5 years (Pigford et al., 1983a). The maximum dose rate from carbon-14, assumed to be in a special carbonate form (Pigford, 1983b), occurs at 2000 years. Neptunium-237 is the next greatest dose contributor, its maximum occurring 2×10^5 years after emplacement. Water flow rates are needed to calculate dose-rate magnitudes.

Similar dose calculations, carried to millions of years, are important in the performance assessment of Sweden's KBS-3 plan for a geologic repository (SKBF, 1983), wherein effects of future climatic changes and glaciation were also considered.

Although the calculation of radiation doses to maximally exposed individuals has been used by the Environmental Protection Agency (EPA) and the NRC as a principal criterion for assessing the environmental releases from nuclear reactors and nuclear fuel-cycle facilities, EPA's proposed standard for geologic disposal adopts a criterion for long-term performance assessment that limits the calculated cumulative release of radionuclides to an "accessible environment", calculated over a time period of 10,000 years. EPA's "accessible environment" is currently interpreted as an underground location 10 km or more from the waste packages. Cumulative releases are easily calculated from the time-dependent concentrations that led to the data in Figure 1, multiplied by flowrates and integrated over time. Scoping calculations summarized in the National Research Council study indicate that EPA's cumulative release limits are likely to be easier to meet than an individual dose criterion for wet-rock repositories, and both requirements are likely to be easily met in some repositories.

CONCLUSIONS

Assessing repository performance requires quantitative prediction of the radionuclide source term. The analysis of diffusive-convective mass transfer provides a predictive tool for long-term performance assessment. The relation of these predictions to laboratory leaching data on solid waste forms must be resolved. NRC release-rate requirements are likely to place special demands on waste packages containing more soluble long-lived radionuclides.

Hydrogeologic transport of dissolved radionuclides is a likely long-term event for wet-rock repositories. Data requirements and calculational details depend upon the form of the performance criterion.

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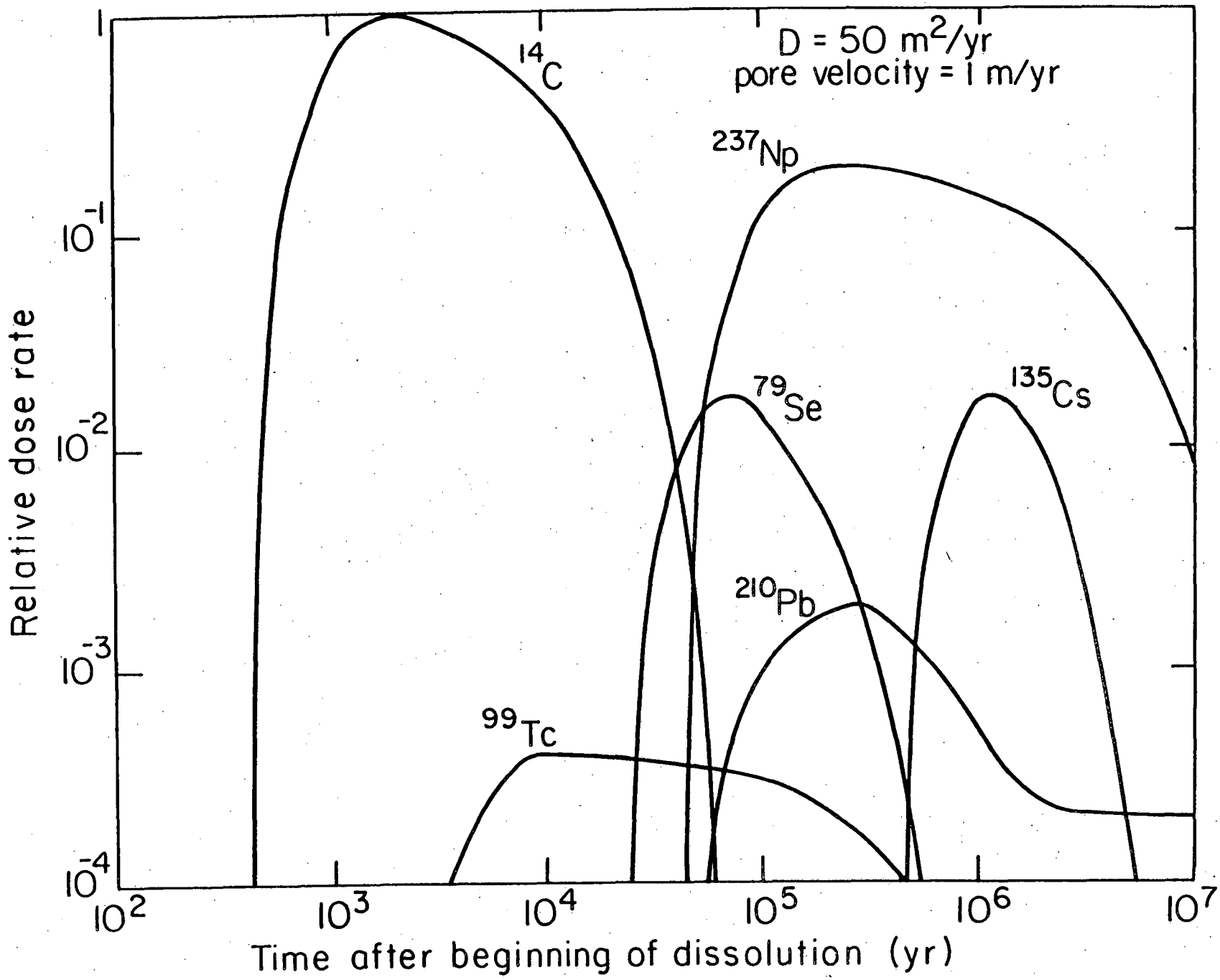

Fig. 1. Relative dose rates at 1000 m from waste.

This report was done with support from the Department of Energy. Any conclusions or opinions expressed in this report represent solely those of the author(s) and not necessarily those of The Regents of the University of California, the Lawrence Berkeley Laboratory or the Department of Energy.

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