

CONF-861102--49



# Lawrence Berkeley Laboratory

UNIVERSITY OF CALIFORNIA

## EARTH SCIENCES DIVISION

Presented at the American Nuclear Society Winter  
Meeting, Washington, DC, November 16-20, 1986

UCI 14 1987

### Mass Transfer of Soluble Species into Backfill and Rock

P.L. Chambre, C.H. Kang,  
W.W.-L. Lee, and T.H. Pigford

July 1986



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

## DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

## DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. Neither the United States Government nor any agency thereof, nor The Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial products process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or The Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or The Regents of the University of California and shall not be used for advertising or product endorsement purposes.

LBL--21807

DE88 000471

Mass Transfer of Soluble Species Into Backfill and Rock

P. L. Chambre', C. H. Kang, W. W.-L. Lee, T. H. Pigford

Department of Nuclear Engineering

and

Lawrence Berkeley Laboratory

University of California

Berkeley, California 94720

In a spent-fuel waste package placed in a geologic repository, the soluble cesium and iodine isotopes accumulated in fuel-cladding gap, voids, and grain boundaries of spent fuel rods are expected to dissolve rapidly when ground water penetrates the fuel rods. Chambre's previous mass-transfer analyses and calculations by Kim, et al.<sup>1</sup> for spent-fuel without backfill show that this soluble "gap activity" of cesium and iodine is the greatest contributor to fractional release rates for these radioelements into surrounding rock, in qualitative agreement with laboratory experiments on spent-fuel rods with locally perforated cladding<sup>2</sup>. Chambre<sup>3</sup> has extended the mass-transfer analysis to include diffusion of soluble species through backfill

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

gsw

into surrounding rock in one-dimensional planar geometry. Here we present numerical illustrations of release rates into porous rock as a function of backfill thickness for parameters typical of a wet-rock repository. Results are compared with the NRC criterion for release rates.

We assume that fuel cladding and a container are not present, water contacts the interior of spent-fuel rods shortly after emplacement, and 1 percent of the total inventory of cesium and iodine is rapidly dissolved into the "void water" that fills voids in the waste package. The void water is equivalent in volume to a 7.4-cm thick layer of water between the waste solid and backfill. Ground-water flow is assumed to be small enough that mass transfer through backfill and into the rock is controlled by molecular diffusion. Time-dependent fractional release rates at the backfill/rock interface, normalized to initial inventories, are shown in Figure 1 for a diffusion coefficient of  $10^{-5}$  cm<sup>2</sup>/s, backfill porosity of 0.2, rock porosity of 0.01, a concentration-based distribution coefficient of 100 for cesium, and for a backfill thickness of 30 cm. Nonsorbing iodine-129 arrives at the backfill/rock interface in less than a year, with a peak release rate about tenfold less than the equivalent fractional release rate limit calculated from the NRC criterion<sup>4</sup>. Cesium-135 and cesium-137 arrive later simultaneously, but the normalized peak release rate of cesium-137 is less because of more rapid decay. The peak release rate of cesium-135 is about tenfold less than its release rate limit,

but the peak release rate of cesium-137 exceeds its limit by several orders of magnitude for hundreds of years.

Figure 2 shows the fractional release rate of cesium-137, normalized to its initial inventory, as a function of time for various backfill thicknesses. Thicker backfills delay arrival at the backfill/rock interface and allow greater decay in the backfill. Backfill several times thicker than in current repository designs would be needed for compliance with the NRC release rate criterion, unless there can be provided a reliable long-lived container with few early failures, much greater sorption in backfill, and/or much lower tortuosity-corrected diffusion coefficients and porosity in backfill and rock.

#### Acknowledgment

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

References

1. C. L. Kim, P. L. Chambre', and T. H. Pigford, "Mass-Transfer-Limited Release of a Soluble Waste Species," Trans. Am. Nuc. Soc., 52, 80, 1986.
2. S. Stroes-Gascoyne, L. H. Johnson, P. A. Beeley and D. M. Sellinger, "Dissolution of Used CANDU Fuel at Various Temperatures and Redox Conditions," in L. Werme (ed.), Scientific Basis for Nuclear Waste Management IX, (1986).
3. P. L. Chambre', To be published.
4. U. S. Nuclear Regulatory Commission, Title 10, Code of Federal Regulations, Part 60, 1983.

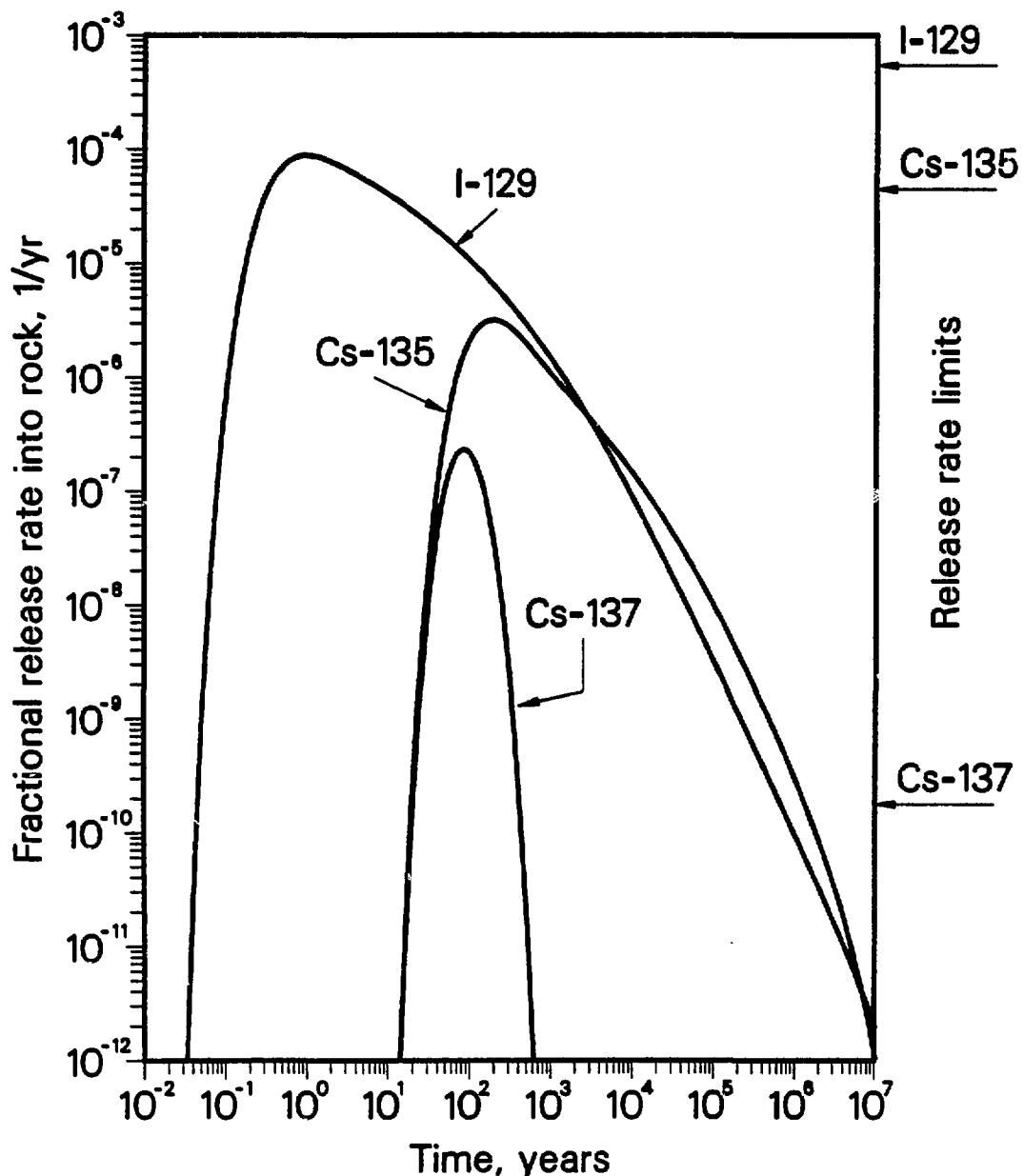


Figure 1. Fractional Release Rates for Various Nuclides Through 30 cm of Backfill

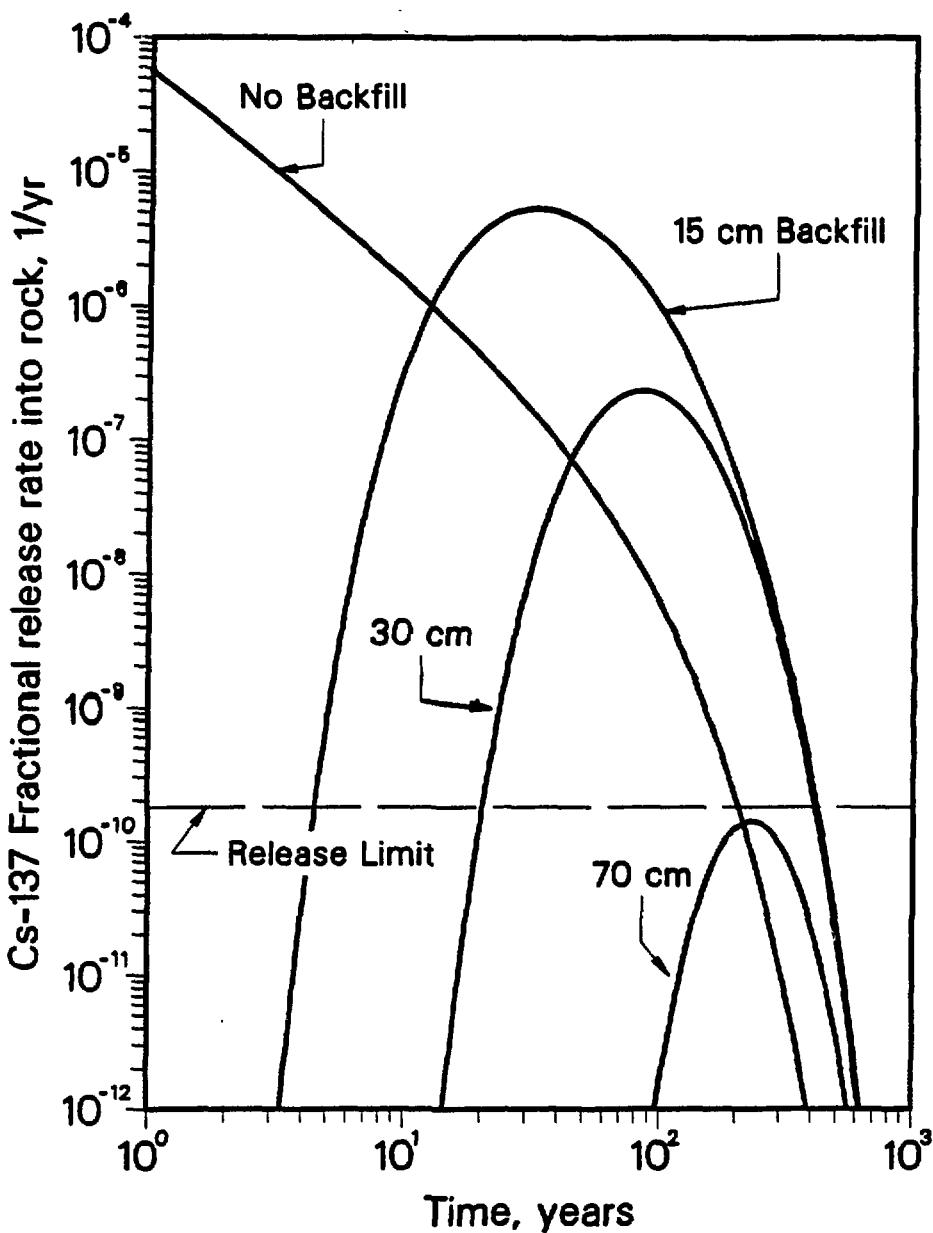


Figure 2. Cesium-137 Fractional Release Rates as a Function of Backfill Thickness and Time