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

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Mass Transfer of Soluble Species Into Backfill and Rock

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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. Chambré'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

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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  $10^{-5} \text{ cm}^2/\text{s}$ , 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

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## References

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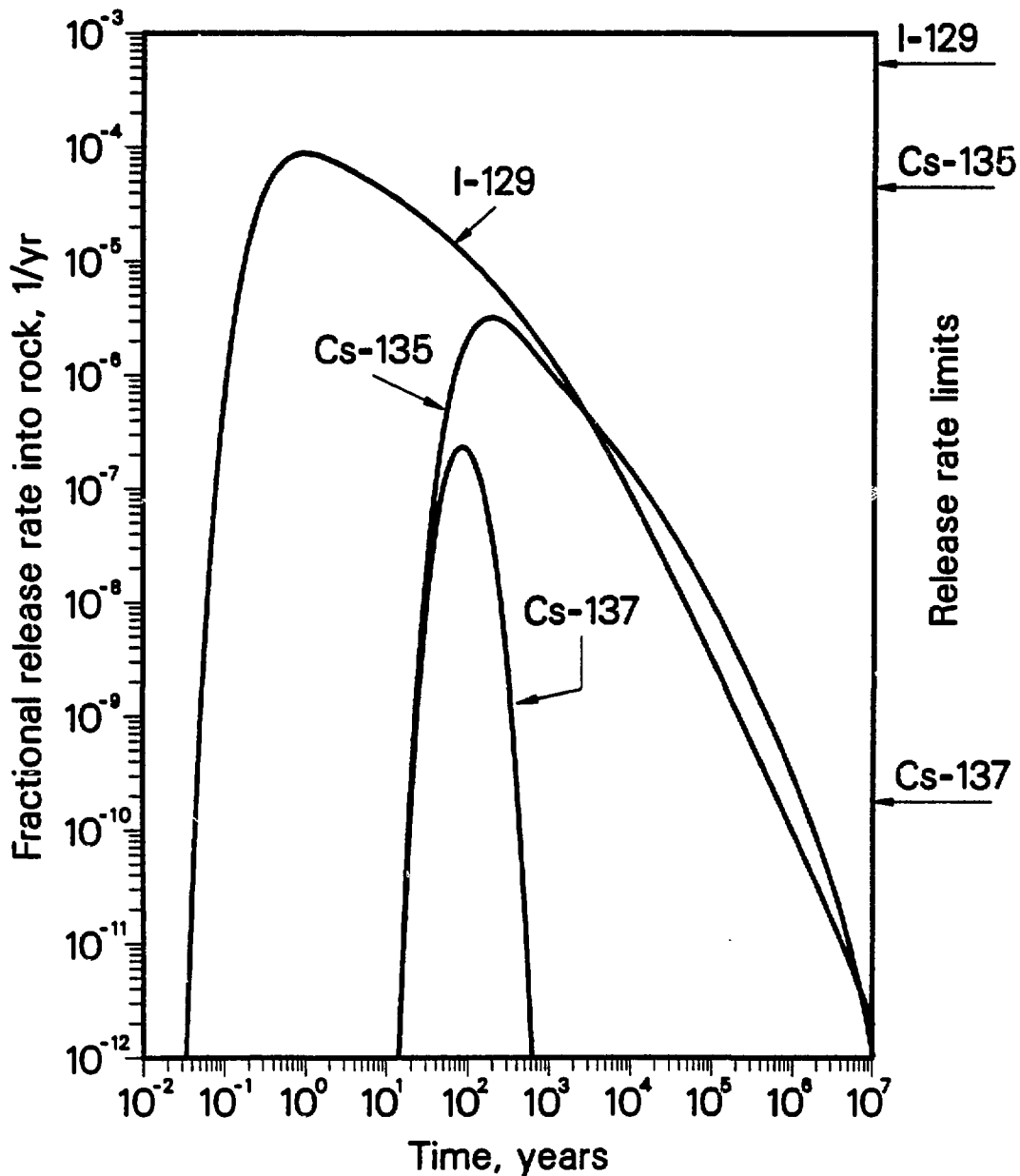


Figure 1. Fractional Release Rates for Various Nuclides Through 30 cm of Backfill

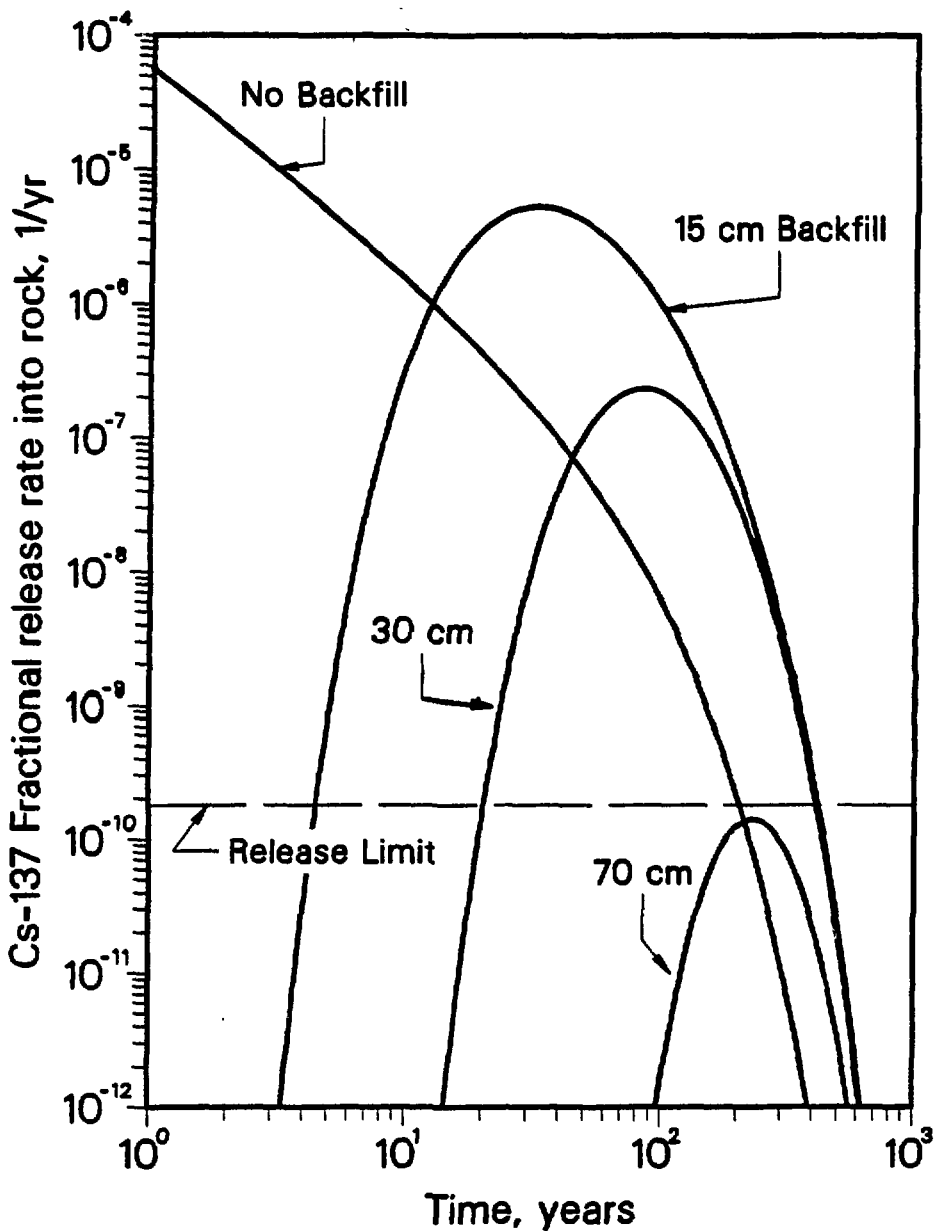


Figure 2. Cesium-137 Fractional Release Rates as a Function of Backfill Thickness and Time