Title

RELEASE RATES OF SOLUBLE SPECIES AT YUCCA MOUNTAIN: A REPLIMINARY MASS TRANSFER ANALYSIS

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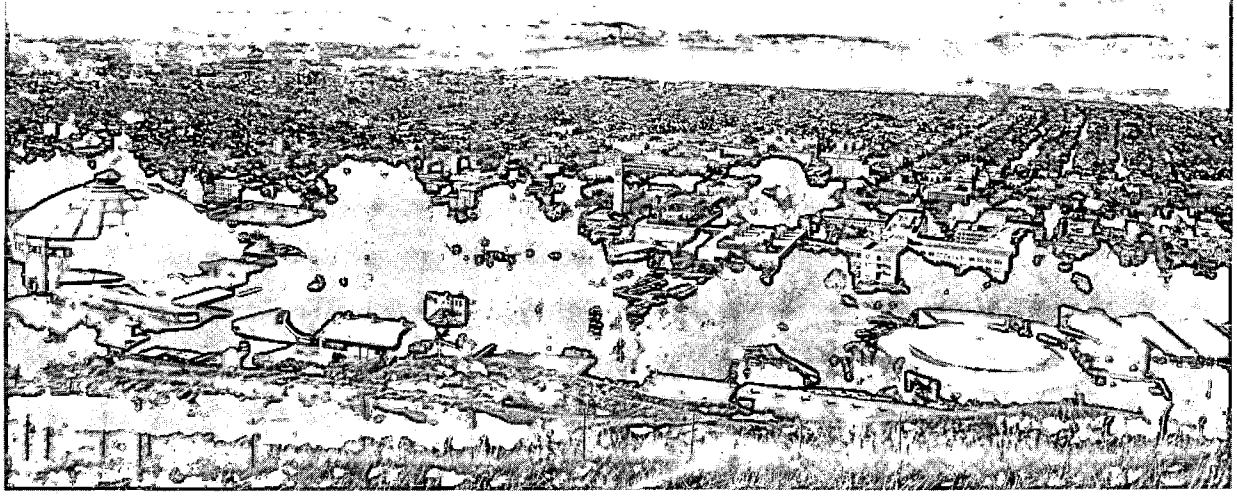

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Release Rates of Soluble Species at Yucca Mountain: A Preliminary Mass Transfer Analysis

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Introduction

To determine compliance with the U. S. Nuclear Regulatory Commission's release rate criterion[1] for the engineered barrier system, the Yucca Mountain project plans to analyse a bounding case which can be summaried as an abundant water and bare waste case.[2] In this technical note we present some preliminary results of using mass-transfer analysis to predict the fractional release rates of soluble species such as ^{135}Cs, ^{129}I and ^{99}Tc.

Analysis

For species that dissolve readily in water, we assume that a specified amount dissolves instantaneously into a water-filled gap or void space in contact with the porous rock. Linear geometry is used. The dissolved species migrate into the porous material under the influence of a concentration gradient. It is expected that advective transport in the pore liquid will be relatively small, so that the governing equation for this migration is

$$K\frac{\partial N(x,t)}{\partial t} = D\frac{\partial^2 N(x,t)}{\partial x^2} - \lambda K N(x,t), \qquad x > a, \quad t > 0 \tag{1}$$

where $N(x,t)$ is the species concentration in the pore liquid,
K is the retardation coefficient, and
D is the diffusion coefficient.
The initial and boundary conditions are

$$N(x,0) = 0, \qquad x > a \tag{2}$$

$$N(a,t) = c(t), \qquad t \geq 0 \tag{3}$$

$$N(\infty,t) = 0, \qquad t \geq 0 \tag{4}$$

where a is the interface between the void space and rock, and
$c(t)$ is the time-dependent of the soluble species in the water in the gap or void water. Because this void space is small, we assume that it is well-mixed and that $c(t)$ is not position-dependent. To solve for $c(t)$, the mass balance in the void is

$$V\frac{dc(t)}{dt} = \dot{m}_f(t) - \dot{m}(t) - \lambda V c(t), \qquad t > 0 \tag{5}$$

where $\dot{m}_f(t)$ is the mass rate of dissolution of the species from the waste form into the void water,
$\dot{m}$ is the mass rate of diffusion into the rock, and
V is the volume of the void water.
To solve (5), we use the initial condition

$$c(0) = c^\circ$$

where c° is the initial concentration of the species in the void water.
The solution below was obtained by Chambré.

$$c(t) = c^\circ e^{-\lambda t}\mathrm{F}(\beta^2 t) + \frac{1}{V}\int_0^t \dot{m}_f(t-\tau)e^{-\lambda\tau}\mathrm{F}(\beta^2\tau)d\tau, \qquad t > 0 \tag{6}$$

where

$$\mathrm{F}(\beta^2 t) \equiv e^{\beta^2}\mathrm{erfc}\sqrt{\beta^2 t}$$

and

$$\beta \equiv \sqrt{DK\epsilon^2/a^2}$$

The mass rate of diffusion of the dissolved species into the tuff is

$$\dot{m}(t) = -SD\epsilon\frac{\partial N(a,t)}{\partial x}, \qquad t > 0 \tag{7}$$

where S is the surface area of the interface between the void space and the tuff. If the void water extends from $x = 0$ to $x = a$ then $S \equiv V/a$. Using (6) the solution to (7) is

$$\dot{m}(t) = N^{\circ}\beta V e^{-\lambda t}\Big\{\frac{1}{\sqrt{\pi t}} - \beta \mathrm{F}(\beta^2 t)\Big\} + \beta \int_0^t \dot{m}_f(t-\tau)e^{-\lambda\tau}\Big\{\frac{1}{\sqrt{\pi\tau}} - \beta \mathrm{F}(\beta^2\tau)\Big\}d\tau, \qquad t > 0 \qquad (8)$$

The fractional release rate of a soluble species whose initial inventory is M_o, denoted f', is

$$f'(t) = \frac{N^{\circ}\beta V e^{-\lambda t}}{M_o}\Big\{\frac{1}{\sqrt{\pi t}} - \beta \mathrm{F}(\beta^2 t)\Big\} + \beta \int_0^t \dot{m}_f(t-\tau)e^{-\lambda\tau}\Big\{\frac{1}{\sqrt{\pi\tau}} - \beta \mathrm{F}(\beta^2\tau)\Big\}d\tau, \qquad t > 0 \qquad (9)$$

Eq. (9) is used to compute fractional release rates for soluble species. The initial concentration c° can be calculated by specifying the void water volume and the amount of the species in the waste that is available for rapid dissolution when water fills the void space.

Numerical Illustrations

We illustrate the above analytic results using conditions typical of a nuclear waste repository at Yucca Mountain. Our reference waste package is reference waste package design No. 4 (3 spent fuel assemblies from pressurized water reactor) as shown on p. 7-28 of the SCP-CD.[2] The fuel assemblies are assumed to be Westinghouse 17 by 17 STD, and the dimensions of the fuel assembly are obtained from p. 2A-343 of DOE/RW-0184.[3] The inventory of various nuclides as a function of time is also taken from the official OCRWM data source, DOE/RW-0184. Table I lists dimensions of the waste package. Table II lists assumed properties of the tuff. Table III lists characteristics of the nuclides used in the calculations.

Table I. Waste Package Dimensions, Spent Fuel from Pressurized Water Reactors

Height	(m)	4.76
Radius	(m)	0.33
Thickness of Gap	(cm)	15.0
Volume of Gap	(m^3)	1.5

Table II. Tuff Properties

Diffusion Coefficient	cm^2/s	10^{-6}
Porosity		0.14

Table III. Characteristics of the Nuclides Studied

	Cs-135	I-129	Tc-99
Retardation Coefficient	9200	1	1
Decay Constant (a^{-1})	2.3×10^{-7}	4.1×10^{-8}	3.3×10^{-6}
Calculated USNRC Release Rate Limit (a^{-1})	5.0×10^{-5}	5.5×10^{-4}	1.0×10^{-5}

The retardation coefficient for cesium was calculated from the sorption ratios in column tests, reported in chapter 4 of the SCP-CD.[2] The porosity value is taken from Bill Glassley's Reference Waste Package Environment report.[4]

We assume that 1%, 10% or 100% of the initial inventories of these soluble species have been released to the fuel-cladding gap, gas plenum, and grain boundaries during reactor operation and dissolve immediately

when water fills the voids in the waste package. For these calculations the slower dissolution from the fuel matrix is neglected. Effects of temperature changes on the diffusion coefficient are also neglected.

Results

In Figures 1, 2, 3 & 4 we show the calculated fractional release rates of ^{135}Cs, ^{129}I and ^{99}Tc to the surrounding tuff as a function of time since the beginning of dissolution. Here the fractional release rate of a species is obtained by calculating the mass release rate and dividing by the 1000-year inventory of that species. Also shown in the figures is the USNRC release rate limit for ^{99}Tc,[1] and the calculated release rate limits for ^{135}Cs and ^{129}I.

The fractional release rate curves for ^{135}Cs, in Figure 1, begin at a higher fractional release and greater negative slope than those for ^{99}T and ^{129}I, because of the greater sorption of cesium in the surrounding tuff. The higher early release rate of cesium depletes the inventory of soluble cesium in the void water and soon results, after a few years, in a cesium fractional release rate lower than that of technetium and iodine. If the readily soluble fraction of cesium is only a few percent, the calculated release limit for ^{135}Cs is exceeded only for a few years, assuming that all waste packages fail at the same time. A distribution of container failures over several decades can result in a repository-average fractional release rate of cesium below the calculated limit, for soluble fractions of a few percent.

For the same readily soluble fractional inventory, the fractional release rate curves for ^{99}T and ^{129}I, Figures 2 and 3, are identical for times up to about 100,000 years, because both species are assumed to be non-sorbing. At later times, the fractional release rates for ^{99}T become smaller because of its shorter half life.

For soluble fractions of a few percent, the release-rate limit for ^{99}T is exceeded for several hundred years, if all containers fail simultanously. Container failures distributed over about a thousand years could result in a repository-average fractional release rate below the regulatory limit.

The calculated release-rate limit for ^{129}I is lower than the limit for ^{99}T, because ^{129}I falls in the regulatory category for a "low-inventory" species. If the readily soluble fraction is only a few percent for iodine, the calculated limit is not exceeded, even if all containers fail at initial placement.

Figure 4 shows the fractional release rates of all three soluble species, if one percent of their respective inventories are dissolved at $t = 0$.

Even if all the inventory of cesium and iodine were readily soluble, the calculated fractional release rates would fall below the calculated limits if the container failures were distributed over about one thousand years.

These calculations are from a bounding analysis that conservatively assumes bare spent fuel, with water in waste-package voids and in contact with saturated porous rock. Because the container, fuel cladding, and corrosion products can still present diffusion barriers after the containers and cladding fail, the more realistic release rates are expected to be less than those calculated here. Calculations of the effects of these additional barriers are in progress.

References

1. U. S. Nuclear Regulatory Commission, Disposal of High-Level Radioactive Wastes in Geologic Repositories, 10 *Code of Federal Regulations* 60.113(a)(1)(ii)(B).

2. U. S. Department of Energy, 1988, *Site Characterization Plan, Yucca Mountain Site,* DOE/RW-0160.

3. U. S. Department of Energy, 1987, *Characteristics of Spent Fuel, High-Level Waste, and Other Radioactive Wastes which may require Long-term Isolation,* DOE/RW-0184.

4. W. E. Glassley, 1986, *Reference Waste Package Environment Report,* UCRL-53726.

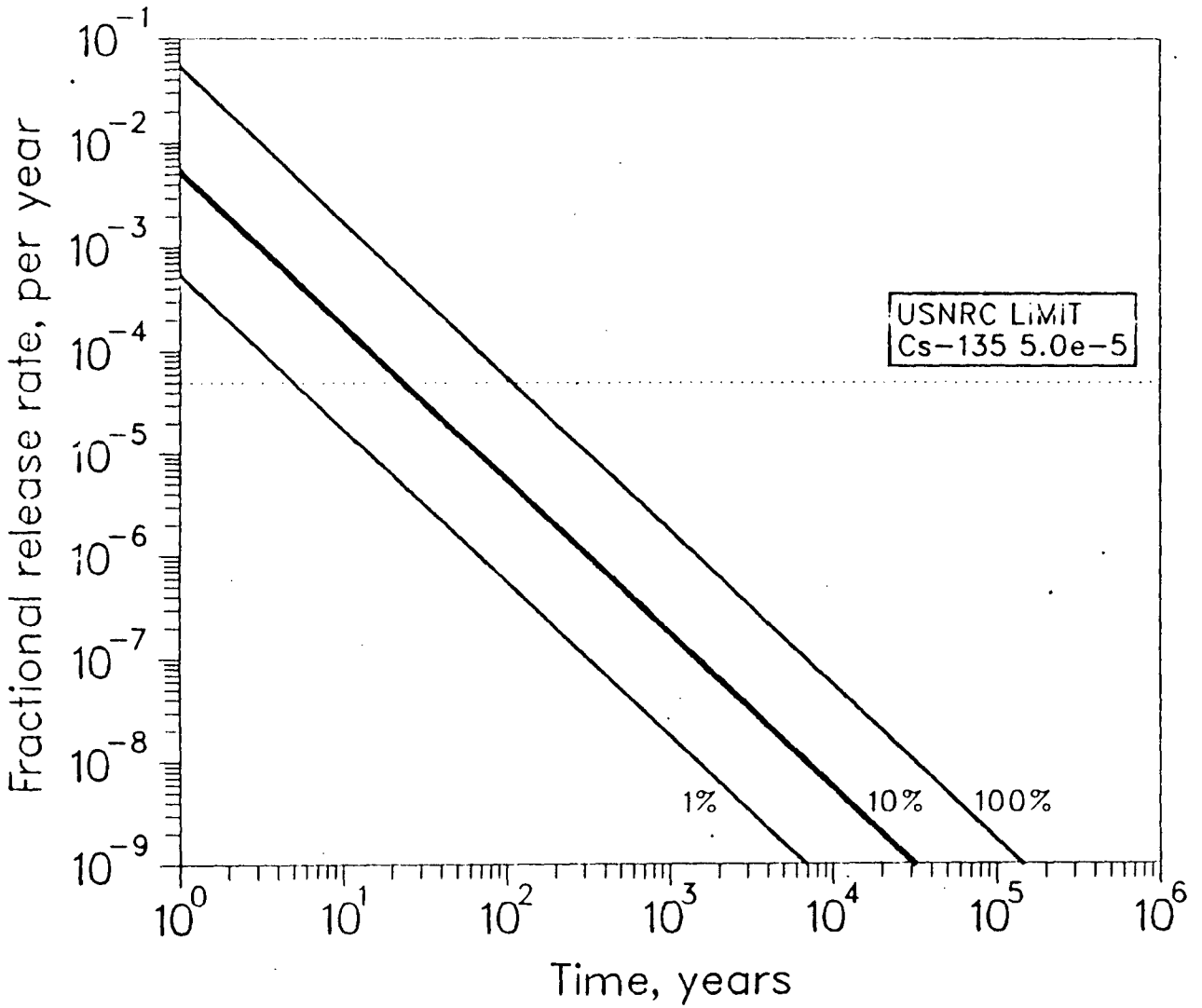

Figure 1. Fractional Release Rates of Cs-135

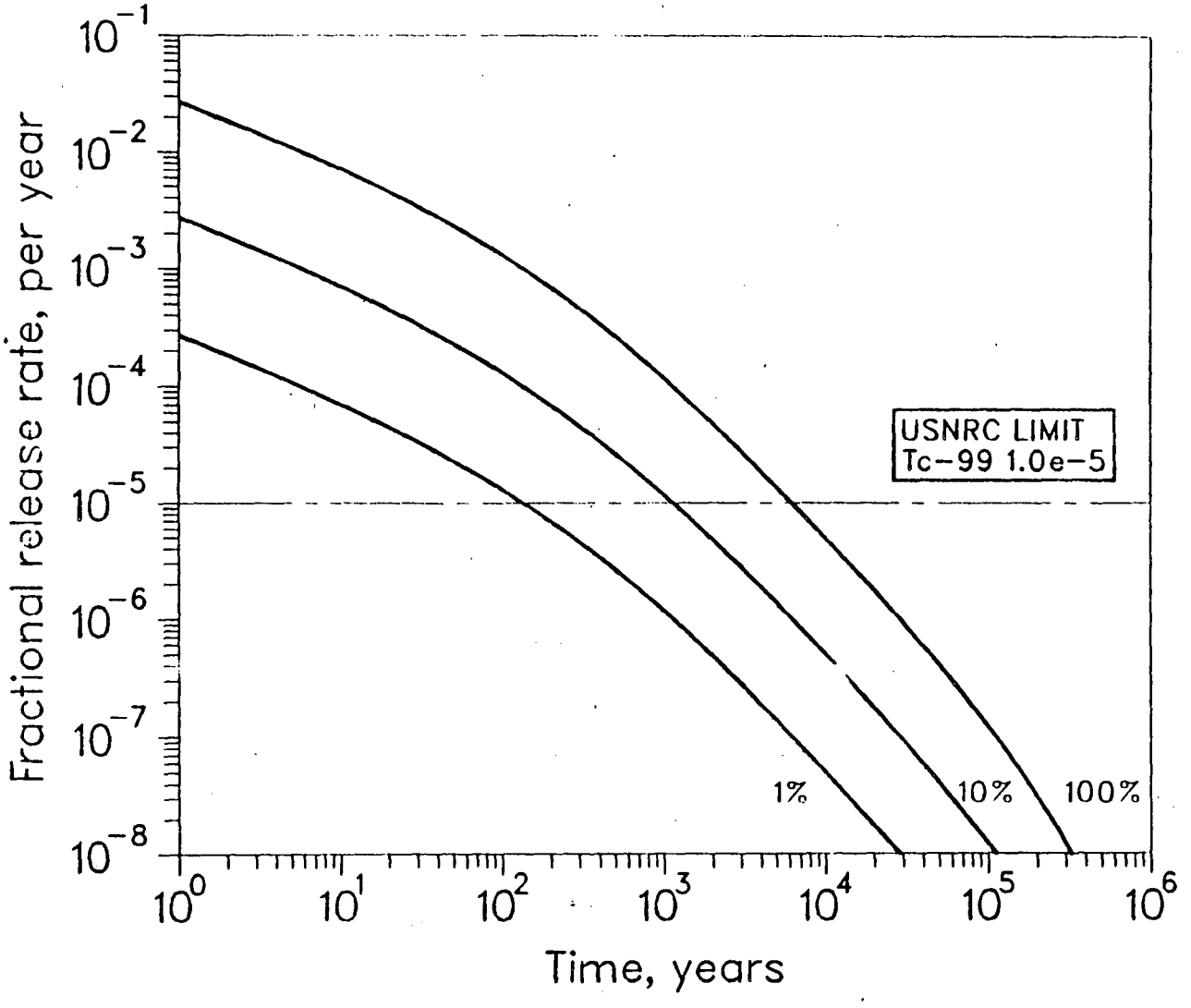

Figure 2. Fractional Release Rates of Tc-99

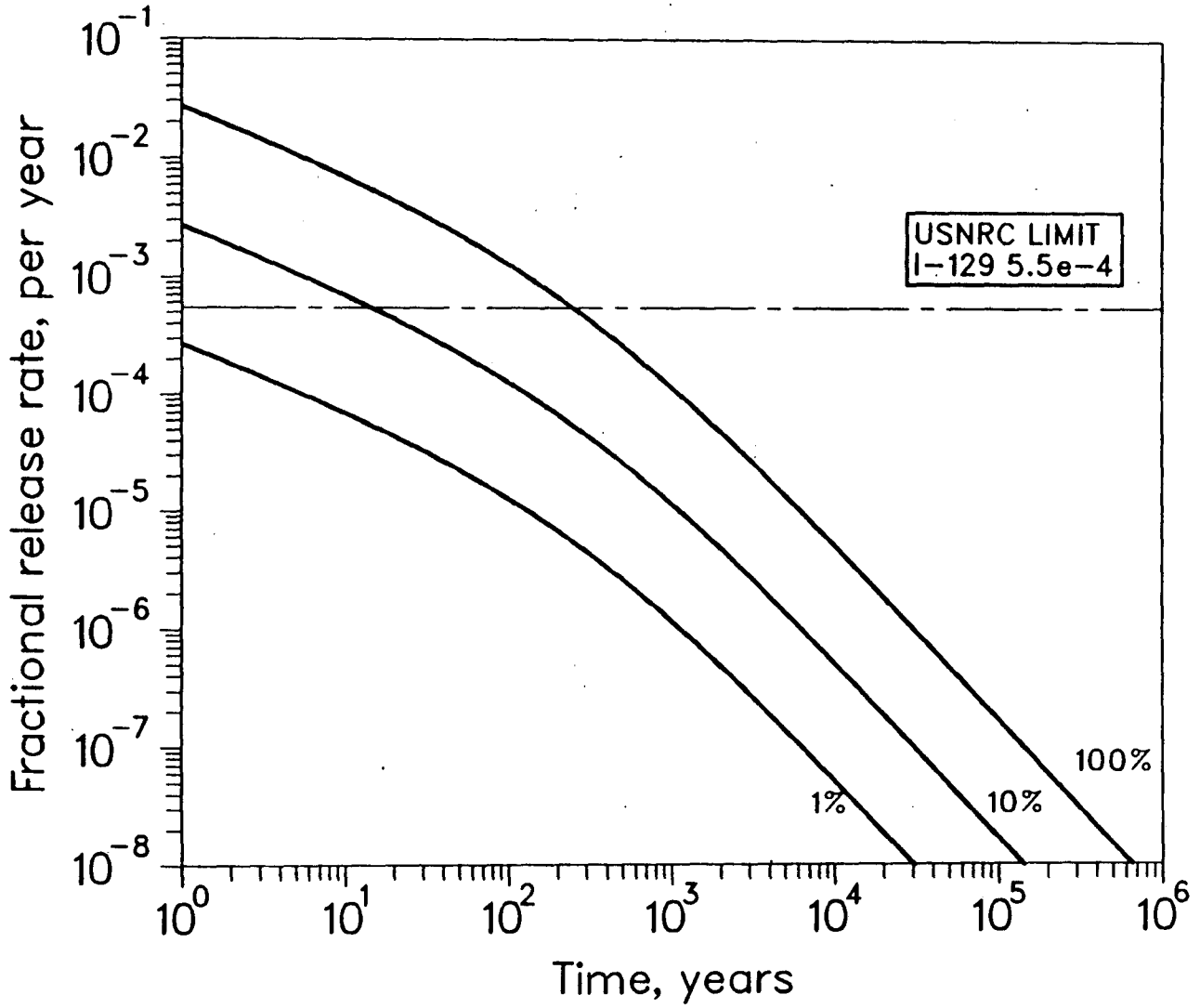

Figure 3. Fractional Release Rates of I-129

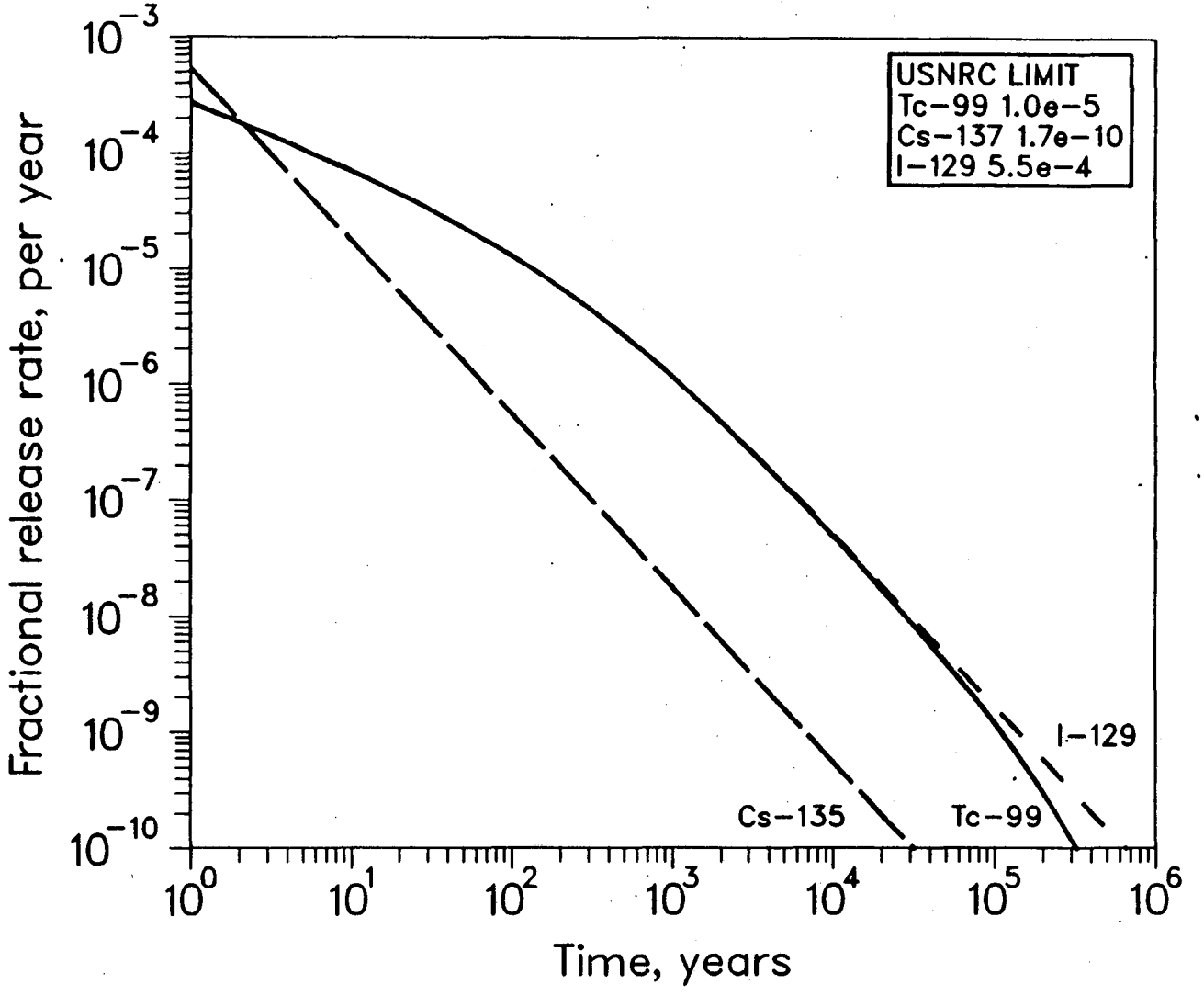

Figure 4. Fractional Release Rates of Soluble Species,1% of inventory dissolves at t=0.

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