

## CRITERIA ASSESSMENT

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### ABSTRACT

A simple analytical model for assessing Federal performance standards is presented. The Criterion Assessment Technique (CAT) model compares repository system performance and current NRC and EPA standards. This paper derives the CAT model and obtains a single parameter that characterizes repository containment capability -- the Retention Time. Conclusions are drawn regarding the relative effectiveness of the Federal standards and the long-term protection afforded by the EPA for individual nuclides.

### INTRODUCTION

The EPA proposed rule 40 CFR 191<sup>1</sup> limits integrated releases of specific nuclides to the accessible environment over  $10^4$  years. The NRC 10 CFR 60<sup>2</sup> restricts release rates from the engineered barrier system. Differences between EPA and NRC criteria lead to questions concerning compatibility and long-term ( $>10^4$  years) effectiveness of these regulations.

The Analytic Sciences Corporation (TASC), under contract to EPRI, has developed a compact model for assessing current Federal performance standards. The model, called the Criterion Assessment Technique (CAT) allows comparisons between repository system performance and existing NRC and EPA standards. The CAT model features a single figure-of-merit for repository containment capability; this capability, the Retention Time, is obtained from a relatively simple analytical model of engineered barrier systems. This paper sketches the derivation of the CAT model and then applies it to the most current EPA and NRC performance standards. Figure 1 depicts a conceptual repository system and the relevant EPA and NRC requirements.

### MODEL OVERVIEW

The principle behind the CAT model is that performance of natural and engineered repository barriers can be described by a decay constant characteristic of the entire system. The overall system Retention Time is found to be sum of individual barrier characteristic times. A system with a large Retention Time is more effective in reducing outflows than a system with a small Retention Time. Retention Times are nuclide specific.

Processes that contribute to individual barrier Retention Times include:

- Waste package dissolution
- Waste form leaching
- Chemical element solubility
- Diffusion in backfill materials
- Hold-up in rock pore volume
- Retardation due to adsorption
- Nuclide migration across the repository length.

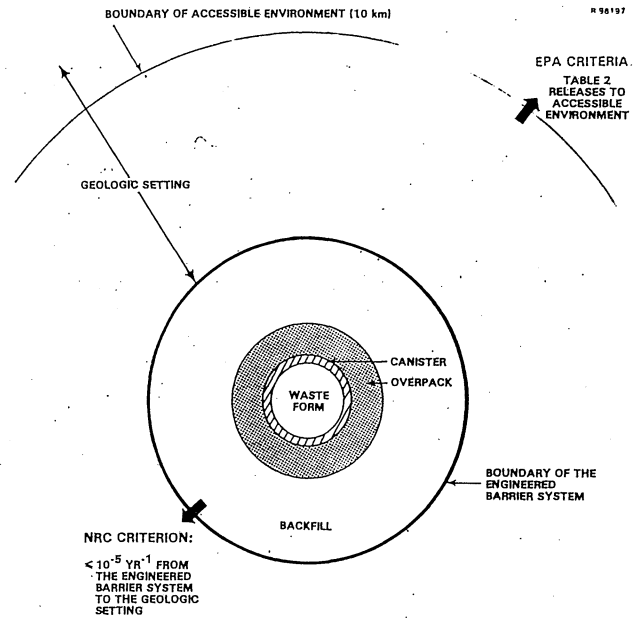


Fig. 1. Conceptual Diagram of Repository System and Regulatory Criteria

### DERIVATION OF CAT MODEL

The waste package consists of reprocessed high-level waste or spent fuel enclosed by a metal overpack. Surrounding the overpack is a buffer region. The buffer material may be a combination of clay and crushed rock designed to inhibit the inflow of water to the waste package and the outflow of dissolved radioactive species. The waste package is located in fractured or porous rock. Water in the vicinity of the waste package flows vertically due to buoyancy forces. Above the waste package is a backfilled storage room with a large pore volume. The objective is to predict the flow of dissolved radionuclides from the storage room, i.e., from the engineered system.

## Solubility and Leaching Model

A reference zero time ( $t = 0$ ) is defined to be the time at which the overpack has substantially corroded and the waste form has begun its process of dissolution.

We consider in the waste package a single nuclide that does not undergo radioactive decay. Let

$m_0$  = Initial nuclide mass at  $t = 0$

$m(t)$  = Mass remaining in undissolved form at time  $t$

$C^*(t)$  = Concentration of nuclide dissolved in water at the surface of the waste package

$C_s$  = Solubility limited concentration of the nuclide in water. Note  $C^*$  cannot exceed  $C_s$ .

Let us now postulate a kinetic model for the rate at which solid dissolves. It is physically reasonable to assume that dissolution rate is proportional to both:

- The amount of nuclide  $m(t)$  in solid form
- The difference in concentration between the solubility limit  $C_s$  and the actual concentration in water  $C^*(t)$

The dissolution model can be expressed as:

$$-\frac{dm(t)}{dt} = Km(t) [C_s - C^*(t)] \quad (1)$$

where  $K$  is a rate constant to be determined.

To estimate  $K$  consider the initial phase of dissolution when the concentration of dissolved chemical species is low. During this period the dissolution can be assumed to be controlled by the bulk leach rate of the waste matrix. This rate is merely the familiar fractional leach rate (denoted as  $L$ ).

$$\text{Thus, } L = -\frac{1}{m(t)} \frac{dm(t)}{dt} \Big|_{t=0} = KC_s \quad (2)$$

$$\text{and } K = L/C_s \quad (3)$$

Equation (1) is non-linear. It can be linearized to give

$$-\frac{dm}{dt} = L[m(t) - \frac{m_0}{C_s} C^*(t)] \quad (4)$$

## Diffusion Across the Buffer Region

Flow of waste across the buffer region is rigorously described by a diffusion equation. Diffusion is driven by a concentration difference between the

two sides of the buffer after failure of the waste package containment. In this simplified model we consider only the steady-state solution of the diffusion equation. The concentration varies linearly with position through the buffer region, and the flow rate can be expressed by:

$$-\frac{dm}{dt} = DA(C^* - C) \quad (5)$$

where  $C$  = Nuclide concentration at the buffer-rock interface

$D$  = The effective buffer diffusion coefficient, and

$A$  = Area of the buffer region normal to the flow.

## Pore Volume of the Storage Room

Flow from the buffer moves rapidly in a vertical direction through the fractured or porous rock between the waste package buffer and the storage room. This storage room is modeled as a "well-stirred" vessel with waste entering from below and leaving from the top of the room. The equation expressing this physical process is

$$-\frac{dm}{dt} = QC + VR \frac{dC}{dt} \quad (6)$$

where  $Q$  = Volumetric flow rate of water through the storage room

$V$  = Pore volume of the storage room

$R$  = Retardation factor associated with nuclide adsorption on the room backfill material.

## Solution of Flow Equations

Equations (4,5 and 6) constitute a linear, second order differential equation:

$$\tau_1(\tau_2 + \tau_3) \frac{d^2C}{dt^2} + (\tau_1 + \tau_2 + \tau_3 + \tau_4) \frac{dC}{dt} + C = 0 \quad (7)$$

where the various  $\tau$ 's are time constants:

$$\tau_1 = VR/Q \quad (\text{holdup}) \quad (8)$$

$$\tau_2 = 1/L \quad (\text{leaching}) \quad (9)$$

$$\tau_3 = m_0/C_s DA \quad (\text{diffusion}) \quad (10)$$

$$\tau_4 = m_0/QC_s \quad (\text{solubility}) \quad (11)$$

The solution to Eq. (7) subject to the initial conditions  $C(0) = 0$  and  $m(0) = m_0$  is

$$\frac{QC}{m_0} = \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} \left[ e^{-\lambda_1 t} - e^{-\lambda_2 t} \right] \quad (12)$$

The quantity  $QC/m_0$  represents the fraction of the initial inventory of any nuclide that flows out of the engineered system per year. The system response constants  $\lambda_1$  and  $\lambda_2$  are found from

$$\lambda_1 + \lambda_2 = \frac{\tau_1 + \tau_2 + \tau_3 + \tau_4}{\tau_1(\tau_2 + \tau_3)} \quad (13)$$

and

$$\lambda_1 \lambda_2 = \frac{1}{\tau_1(\tau_2 + \tau_3)} \quad (14)$$

Equations (13) and (14) would involve additional time constants if other physical processes were modeled.

#### Radioactive Decay

It can be shown from Eqs. (4, 5 and 6) that including radioactive decay results in a simple modification of Eq. (12). For the simple exponential decay case, the right-hand side of Eq. (12) is multiplied by the factor  $e^{-\lambda_d t}$ , where  $\lambda_d$  is the radioactive decay constant.

#### Approximation of the Fractional Flow Pulse Shape

The fractional flow equation, Eq. (12), contains two parameters,  $\lambda_1$  and  $\lambda_2$ , that describe the physical characteristics of the engineered repository. To judge the relative effectiveness of the engineered system in meeting NRC and EPA performance regulations, it is necessary to focus on two important characteristics of the flow equation adjusted for decay, i.e.,

$$\frac{QC}{m_0} = \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} \left[ e^{-\lambda_1 t} - e^{-\lambda_2 t} \right] e^{-\lambda_d t} \quad (15)$$

These characteristics are (1) the peak flow rate and (2) the integrated flow from time of emplacement to  $10^4$  years after emplacement.

A suitable approximation to Eq. (15) is sought that describes the nuclide pulse shape by a single parameter rather than by two parameters. The approximation should provide reasonable upper bounds to both the peak and integrated flow rates. One simple approximate form is:

$$\frac{QC}{m_0} = \gamma^2 t e^{-(\gamma + \lambda_d)t} \quad (16)$$

where

$$\gamma = e \frac{\lambda_1 \lambda_2}{\lambda_1 + \lambda_2} \quad (e = 2.718) \quad (17)$$

Its characteristics are:

$$f_p = \frac{\gamma^2}{(\gamma + \lambda_d)e} \quad (18)$$

where  $f_p$  is the peak fractional release, and

$$f_i = \frac{\gamma^2}{(\gamma + \lambda_d)^2} \left[ 1 - [1 + (\gamma + \lambda_d) T] e^{-(\gamma + \lambda_d)T} \right] \quad (19)$$

where  $f_i$  is the integrated fraction released;  $T$  is the elapsed time from waste package degradation up to  $10^4$  years. If overpack failure occurs at  $2 \times 10^3$  years after emplacement, for instance, then  $T = 8 \times 10^3$  years.

That both Eqs. (18 and 19) provide conservative estimates for  $f_p$  and  $f_i$  can be verified by performing similar operations on Eq. (15). These approximations are also reasonable ones when  $\lambda_1$  and  $\lambda_2$  are not widely different, as is the case in repository systems examined to date. Use of Eq. (16) in place of Eq. (15) results in errors less than a factor two in  $f_p$  and  $f_i$ .

#### Retention Time

The characteristic repository response parameter  $\gamma$  may be determined in terms of the response times  $\tau_1$  through  $\tau_4$ . The result is

$$\gamma = \frac{e}{\tau_1 + \tau_2 + \tau_3 + \tau_4} \quad (20)$$

The sum of the individual barrier time constants  $\tau_1$  through  $\tau_4$  has been identified in a previous study for EPRI<sup>34</sup> as a critical system parameter. It has been designated as the repository system Retention Time,  $\tau_{sys}$ . Thus,

$$\gamma = \frac{e}{\tau_{sys}} \quad (21)$$

The following discussion of Federal criteria is general and does not depend upon any particular selection of  $\tau_{sys}$ . The Retention Time or the parameter  $\gamma$  may, however, be determined for a given repository system of barriers from the preceding equations.

#### ASSESSMENT OF FEDERAL CRITERIA

The NRC requires a release rate less than  $10^{-5}$  per year for significant nuclides based on the inventory  $10^3$  years after closure. If the inventory  $m_0$  is evaluated at  $10^3$  years, then the NRC rule would impose

$$f_p \leq 10^{-5} \text{ yr}^{-1} \quad (22)$$

The EPA requires that cumulative releases of nuclides be less than values tabulated in 40 CFR 191 in units of curies per  $10^3$  MTHM. The EPA standard applies strictly to releases at the "accessible environment," which may be kilometers from the boundary of the engineered repository. For present purposes we assume, conservatively, that no attenuation of the nuclide release pulse occurs between the repository and the "accessible environment." This assumption allows the use of our present release model to evaluate the EPA standard.

For any nuclide, let

$I_0$  = The normalized inventory (Ci/10<sup>5</sup> MTHM) at the reference time

R = The stipulated EPA release limit (Ci/10<sup>5</sup> MTHM).

The EPA criterion can be written as

$$\frac{I_0 f_i}{R} < 1 \quad (23)$$

For mixtures of nuclides, the EPA mandates that the  $I_0 f_i/R$  values summed over all nuclides should be less than one. Considering the small number of potentially hazardous nuclides and the large uncertainties in performance projections, use of Eq. (23) for individual nuclides cannot be in serious error.

#### DOSE RATE ESTIMATES

Peak individual dose rates resulting from a peak fractional release, Eq. (18), can be estimated using the  $I_0/R$  ratio. A release of R curies of any nuclide should, according to the EPA environmental model, result in 10<sup>3</sup> health effects. Assume, in addition, that each health effect corresponds to roughly 5×10<sup>3</sup> man-rem<sup>4</sup> and that the affected population is of the order of 10<sup>6</sup>. For these assumptions the dose rate, D, can be determined from

$$D = 5000 \frac{I_0}{R} f_p \quad (\text{mrem/yr}) \quad (24)$$

#### COMPARISON OF FEDERAL CRITERIA

Any nuclide can be characterized by its normalized inventory  $I_0/R$  and its half-life  $T_{1/2} = 0.693/\lambda_d$ . Any repository system can be characterized by  $\tau_{\text{sys}}$ , its Retention Time. The solid curves in Fig. 2 represent the Retention Time requirements for the repository needed to meet the EPA criterion as a function of  $I_0/R$  and  $T_{1/2}$ .  $T$  is assumed to be 8×10<sup>3</sup> yr in the example. Increasing inventories and increasing half lives mandate larger Retention Times.

Shown in Fig. 2 as a dashed curve is the NRC criterion. To use this criterion choose a nuclide half-life and interpolate the requisite  $\tau_{\text{sys}}$  value from the ordinate of the dashed curve. Note the NRC criterion is independent of  $I_0/R$ .

Ten individual nuclides selected because their  $I_0/R$  values exceed one are indicated at appropriate  $I_0/R$ ,  $T_{1/2}$  coordinates in Fig. 2. Interpolation between  $\tau_{\text{sys}}$  curves gives the repository system Retention Time needed to meet EPA standards. Nuclides located below the dashed (NRC) curve will require yet larger Retention Times to meet NRC standards. Spent fuel inventories were used to assign nuclide

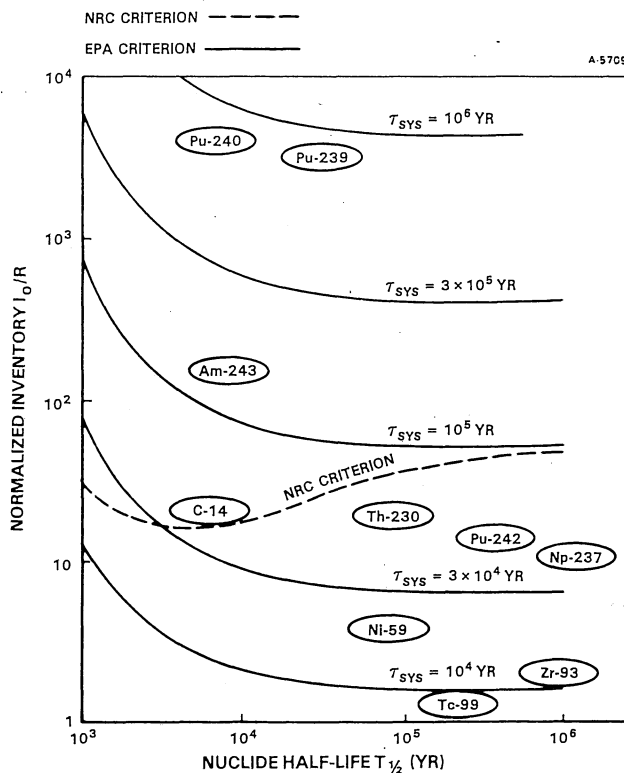


Fig. 2. Comparison of Federal Criteria

$I_0/R$  values. A special case is Th-230, whose inventory was very conservatively assumed to be that of its precursor, U-234. Th-230 may also be used to infer upper bound requirements on its daughter nuclide Ra-226.

The following observations can be made from Fig. 2:

- For nuclides with high hazard potential, i.e., high  $I_0/R$  (C-14, Pu-239, Pu-240 and Am-243), the NRC limit is easier to meet than the EPA limit. Above the dashed curve, the EPA limit controls
- For nuclides with low  $I_0/R$  (Th-230, Pu-242, Np-237, Ni-59, Tc-99 and Zr-93) the EPA standard is easier to meet. Below the dashed curve, the NRC limit controls
- Actinides generally require higher Retention times; however, owing to low solubility and high retardation, Retention Times exceeding 10<sup>6</sup> yr are in practice attainable for these nuclides. Demonstrating the needed 5×10<sup>4</sup> yr retention for C-14 could be a problem in certain repository systems.

Figure 3 shows lines of equal dose rate on an  $I_0/R, T_{1/2}$  plot. The solid curves represent the peak dose rates to typical individuals (at any time after closure) when the EPA limit is exactly met. The horizontal dashed isodose rate lines apply when the NRC standard is exactly met. The coordinates of the ten individual spent fuel nuclides are also depicted as in Fig. 2. Note that two 1 mrem/yr EPA isodose rate curves exist; between them occurs a minimum dose rate of about 0.8 mrem/yr.

It may be concluded from Fig. 3 that:

- Meeting EPA standards for all nuclides results in a peak individual dose rate at or below 2 mrem/yr. The isotope Pu-239 is the worst in this regard
- For nuclides for which the NRC criterion controls Retention Time requirements (low  $I_0/R$ ), peak dose rates are simultaneously limited to values at or below 2 mrem/year by the EPA criterion
- When the EPA is controlling (high  $I_0/R$ ) it does so by relatively high margins. Meeting NRC standards could result in greater than 100 mrem/yr dose rates.

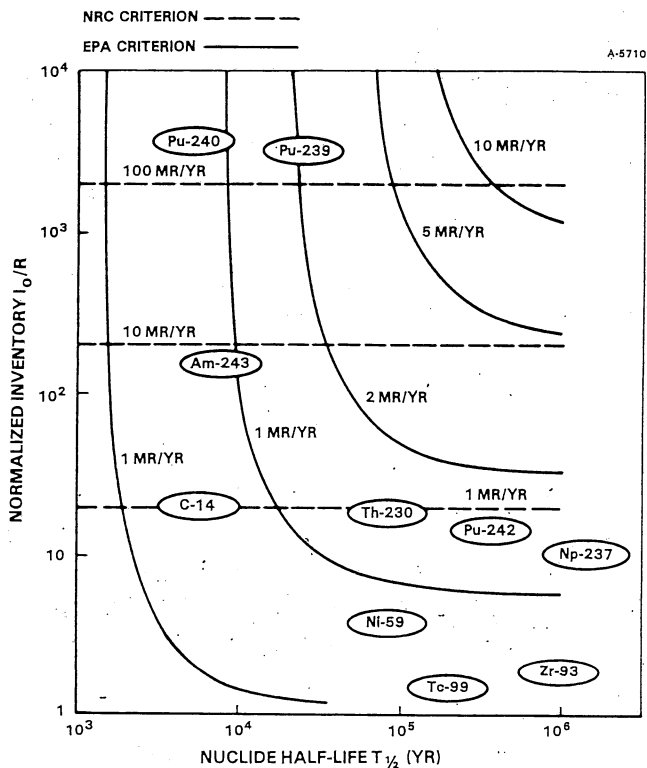


Fig. 3. Individual Dose Rates from Federal Criteria

In practical terms, the NRC fractional release rate criterion does not play a key role in assuring public health and safety for HLW disposal. It does, however, provide a supplemental margin of confidence.

#### CONCLUSIONS

Results indicate that Retention Times on the order of  $10^4$  to  $10^5$  yr are required for compliance with NRC and EPA criteria. Certain actinides do require longer retention in the engineered system, but actual Retention Times are generally greater by one or more orders of magnitude than those necessary to meet criteria. It is found that meeting EPA criteria does ensure safety to individuals by limiting peak dose rates at times in excess the  $10^4$  year regulatory period. This conclusion assumes the EPA environmental model (R values) is reasonably correct. Furthermore, no case was encountered in which the NRC release rate standard was necessary to provide safety.

#### REFERENCES

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