**ABSTRACT**

Partially failed containers may provide a meaningful barrier to the release of gaseous radionuclides. A modeling approach is outlined and sample calculations are provided that show the effects on release due to a limited perforation area, to decreasing temperature, and to the partial occlusion of the perforated area by corrosion products.

**INTRODUCTION**

Under disposal conditions in the unsaturated zone, the reference condition in the United States, $^{14}\text{C}$ can be released from the spent fuel as soon as a container is breached because of its presence in the cladding waste. The most important mechanism for $^{14}\text{C}$ mobilization is its oxidation, in air, into gaseous $^{14}\text{CO}_2$. From the latest version of the Yucca Mountain Site Characterization Plan (SCP), the following prompt oxidation fractions, $f$, of the total $^{14}\text{C}$ inventory within the spent fuel can be deduced:

- $f = 1\%$ for $T > 215^\circ\text{C}$
- $f = 0.2\%$ for $170 < T < 215^\circ\text{C}$
- $f = 0.1\%$ for $T < 170^\circ\text{C}$

These prompt oxidation fractions have to be augmented further, by adding $2\%$, at least, in order to reflect recent experimental data under both oxidizing and "inert" conditions.

The most stringent criterion on waste package (WP) performance promulgated by the U.S. Nuclear Regulatory Commission (NRC) applies when the WP is at its hottest and stipulates that, for a period of 300 to 1,000 years, the WP must provide a "substantially complete containment" of all the radionuclides that have been disposed of. Although "substantially complete containment" is a term of law rather than a term of science, it needs to eventually be quantified, as 100% absence of containers failure can never be demonstrated. We assume, therefore, that a certain number of containers is allowed to fail provided that the $^{14}\text{C}$ annual release from the ensemble of spent fuel containers is confined to $10^{-6}$ of the total inventory of $^{14}\text{C}$ present in any given year. This is a more restrictive criterion than the analogous $10^{-5}$ criterion which applies after the substantially complete containment period.

The SCP suggests that the $^{14}\text{CO}_2$ is released instantly from the breached container. Under this assumption, exceedingly few containers (1 in 25,000 roughly) can fail per year even under the $10^{-6}$ criterion. It can be argued, however, that prompt oxidation of $^{14}\text{C}$ into $^{14}\text{CO}_2$ should not necessarily result in prompt release of all the oxidized fraction out of the spent fuel containers unless the average perforation area in the container surface is large enough. In other words, a small average perforation area with respect to the total surface area of the container could still guarantee that the release is slow enough to allow more reasonable container failure rates. Our initial calculations support this point.

*Roughly 38% of the $^{14}\text{C}$ in spent fuel resides in the UO$_2$ pellets; 62% is associated with the rest of the assembly materials (cladding waste).*
MODELING FORMALISM

Upon performing a $^{14}$CO$_2$ mass balance within a single container, the following equation can be written in terms of $N(t)$, the number of $^{14}$CO$_2$ molecules in the empty volume within the container:

$$\frac{dN}{dt} = -[J_d(t) + J_c(t)]A(t) - \lambda N(t) + S(t)$$

(1)

where $J_d(t)$ represents the diffusive mass flux away from the container, $J_c(t)$ is the convective mass flux, $A(t)$ is the area of the penetration as it evolves with time, $\lambda N(t)$ is the removal rate due to decay, and $S(t)$ is the production rate of $^{14}$CO$_2$ due to oxidation of the $^{14}$C.

Ideally, the rate of $^{14}$C oxidation into $^{14}$CO$_2$ is dependent on the rate of supply of O$_2$. We assume, conservatively, that as soon as the perforation penetrates the container all the oxygen needed to oxidize the $^{14}$C is instantly available and instantly replenished. One can assume several $^{14}$CO$_2$ production rate functions. Instant oxidation at time $t=t_0$ is modeled as follows:

$$S(t) = G(T) \delta(t-t_0)$$

(2)

where the function $\delta(t)$ represents Dirac's delta and T the temperature, itself a function of time. Alternatively, if one wants to model a prompt oxidation step followed by a decaying exponential tail, one may assume that:

$$S(t) = S_o \exp[-\alpha(T)(t-t_0)] U(t-t_0)$$

(3)

with $U(t)$ the Heaviside step function and $\alpha$ an experimental constant.

The diffusive term, $J_d(t)$, can be obtained from the literature. The steady-state, diffusive release of a solute per unit area from a cylindrical hole into an infinite medium can be represented as follows:

$$J_d(t) = \frac{D(T) N(t)}{V \cdot (\bar{l} \cdot E^{-1} + 0.25 \cdot [\pi A(t)]^{1/2})}$$

(4a)

where:

- $D(T)$ = temperature-dependent diffusion coefficient
- $V$ = gas volume within the container
- $E$ = open porosity of the hole
- $\bar{l}$ = container wall thickness (penetration length)
- $A(t)$ = hole perforation area as function of time

Usage of the above expression for the diffusive flux assumes that the concentration profile within the cylindrical penetration adjusts itself instantly to a steady-state type profile. This assumption is justified a posteriori noting that the diffusion coefficient is large enough to assure rapid establishment of a steady-state profile.

Since $\bar{l}=1$ cm in Eq.(4a), the dependence of $J_d$ on $A$, the hole perforation area, is negligible as long as the following inequality is satisfied:

$$A < 5.1 \epsilon^{-2}, \ [A]=[cm^2].$$

(4b)

This is likely the case in the applications. Combining the latter observation with the fact that, for non-solubility-limited release, holes next to each other on a container surface do not interact significantly with each other, one can model the release from multiple holes as if it occurred from a single, equivalent hole with the same combined surface area or the equivalent diameter.

At the present stage of our study, we have yet to model the convective flux contribution. Under normal conditions the $\Delta P$ of $^{14}$CO$_2$ across the waste container is negligible as it is confined to values within $5\times10^{-3}$ atm. A proper expression for $J_c(t)$ will be necessary, however, in order to establish the importance of container "breathing" due to (a) pressure variations in the outside environment and (b) occasional pressure surges due to failing fuel rod cladding. Since in both cases the potential release should be proportional to the amount of $^{14}$CO$_2$ present in the container, it can be argued that a general expression for $J_c(t)$ is of the form:

$$J_c(t) = F_c(T) N(t),$$

(5)
with \( F_c(T) \) a function yet to be defined.

The general solution to Equation (1) is then:

\[
N(t) = e^{-\int_0^t H(s) \, ds} \left( \int_0^t e^{\int_0^s H(u) \, du} \, ds \right) \text{dt} + \text{constant}
\]  
\( 6 \)
in which the value of the arbitrary constant is determined by the initial conditions to the problem and:

\[
H(T,t) = \lambda + (F_D + F_C) A(t)
\]  
\( 7 \)
where the term \( F_D \) is simply equivalent to \( J_D/N(t) \) in Eq.(4a).

Equation (6) can be substituted into Equations (4a) and (5) to obtain the rate of release per unit area of the perforation area. The latter expressions can then be integrated to obtain cumulative releases.

At this time, we have produced two analytical solutions\(^7\) to Equation (1). In both cases we do not model convection (\( F_C=0 \)) and we assume (a) isothermal conditions and (b) a fixed size perforated area forming at time \( t=0 \). One solution models the prompt oxidation of the \(^{14}\text{C} \) into \(^{14}\text{CO}_2 \) according to Equation (2); the other solution models prompt oxidation followed by an additional, exponentially decaying tail.

**NUMERICAL RESULTS**

We have produced sample calculations to address the question: what the average perforation area should be in spent fuel containers so that 0.1% (i.e., 25 containers in 25,000) can fail simultaneously in a year and still meet a \( 10^{-6} \) release limit per year from the repository during the substantially complete containment period.

In order to address that problem, we have subdivided the repository into different temperature zones with \( T_{av}=250 \), \( T_{av}=190 \), \( T_{av}=150 \), and \( T_{av}=80^\circ \text{C} \), respectively. For each temperature regime we have assessed the influence on release due to crud potentially occluding part of the perforation area (the higher the degree of occlusion, the smaller the value of the open porosity parameter). We have assumed also a prompt oxidation fraction of 2% plus the SCP suggested fraction. Then we have allowed an extra oxidation fraction of up to 1% during the first 15 days after breach.

Some of our results are reported in Tables 1 and 2.

Table 1 is interpreted as follows: if the average perforation area is less than \( 0.18 \text{ mm}^2 \), the \( 10^{-6} \) criterion is met even if (a) up to 0.1% of the containers fail simultaneously, (b) there is no crud in the hole (\( \epsilon=1 \)), and (c) up to 4% of the total, original \(^{14}\text{C} \) inventory oxidizes (3% promptly + 1% in the tail within two weeks). Lower temperatures or higher degree of hole occlusion (\( \epsilon < 1 \)) would allow larger perforated areas. Indeed (Table 2), the average perforation area becomes as large as \( 613 \text{ mm}^2 \) for \( T_{av}=80^\circ \text{C} \) and \( \epsilon=0.01 \).

In these calculations the diffusion coefficient of \(^{14}\text{CO}_2 \) in air was taken as the same as that of \(^{14}\text{CO}_2 \) in nitrogen,\(^8\) namely:

\[
D(T) = 0.0245 \cdot T^{1.75}, \quad [D]=\text{[m}^2/\text{yr}], \quad [T]=\text{[}^\circ\text{K}].
\]  
\( 8 \)
The gas volume within the container was taken to be \( 1 \text{ m}^3 \). Tortuosity, \( \tau \), was taken into account as function of open porosity, \( \epsilon \), as follows:

\[
\tau = 1 - 4\cdot\log_{10}(\epsilon).
\]  
\( 9 \)

It is to be noted that if no credit were taken for the slowing down of the \(^{14}\text{C} \) release by the hole(s), the allowable container failure rate would have been at least one order of magnitude smaller.

**CONCLUSIONS**

Our preliminary analysis supports the assumption that partly failed containers may offer a significant delay to gaseous \(^{14}\text{C} \) release both during the substantially complete containment and the controlled release periods. A properly implemented approach to obtain credit for this effect may decrease the strict requirement on container failure rates that the present waste package licensing strategy demands.

So far, our analysis can model (a) the finite amount of \(^{14}\text{C} \) present in the spent fuel container, (b) container wall
thickness, (c) perforated area equivalent diameter and growth rate, (d) degree of occlusion of the perforation due to corrosion products, (e) thermal oxidation rate of the $^{14}$C into $^{14}$CO$_{2}$, (f) gaseous diffusion of the $^{14}$CO$_{2}$ in the (partly occluded) hole(s), and (g) radioactive decay. All rate processes can be implemented with their temperature dependence. Further work is needed to incorporate "breathing" of the container caused by barometric pressure variations in the outside environment and the effect of occasional pressure surges due to failing fuel rods. Once these analyses are completed, the relevant expressions for $^{14}$C release should be convoluted with the container breach rate distribution functions that are being developed within the waste management community. One such analysis will identify the allowable container perforation area in order to meet the applicable performance criteria.

The results of the $^{14}$C release analyses from failed containers could be also verified via gas diffusion experiments, which are amply within reach of available technology.

The approach proposed here for gaseous radionuclide release can also be proposed for the release of radionuclides potentially leached by groundwater infiltrating a breached container. The likely presence of crud in container penetrations may allow only very small releases regardless of radionuclide solubilities.

Tables 1 and 2 suggest that, if no earlier penetrations have occurred, one could still tolerate a 0.1% failure of all containers (i.e., 25 containers in the hypotheses of the problem indicated in the Introduction) in a given year provided the equivalent diameter of the average perforated area per container is within the calculated limits. These limits vary from half a millimeter to about 28 millimeters depending on temperature, the amount of crud present in the perforation, and the $^{14}$C oxidized fraction. The presence of crud partly occluding the perforation is an important component of our models.

REFERENCES


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Table 1. Allowable average perforation area [mm²] in order to meet a 10⁻⁶ release/year criterion assuming 0.1% of the containers fail simultaneously.

<table>
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<th>$\varepsilon$</th>
<th>$\Delta f$</th>
<th>0.05%</th>
<th>0.1%</th>
<th>1%</th>
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<td>161.5</td>
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</table>

$T_{av} = 250^\circ C$; prompt oxidation fraction = 3%

Table 2. Allowable average perforation area [mm²] in order to meet a 10⁻⁶ release/year criterion assuming 0.1% of the containers fail simultaneously.

<table>
<thead>
<tr>
<th>$\varepsilon$</th>
<th>$\Delta f$</th>
<th>0.01%</th>
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</tbody>
</table>

$T_{av} = 80^\circ C$; prompt oxidation fraction = 2.1%

$\Delta f$ - further oxidation fraction over 15 days;
$\varepsilon$ - open porosity of the hole.

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