A GENERALIZED DEFINITION FOR WASTE FORM DURABILITY

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ABSTRACT

When evaluating waste form performance, the term "durability" often appears in casual discourse, but in the technical literature, the focus is often on waste form "degradation" in terms of mass lost per unit area per unit time. Waste form degradation plays a key role in developing models of the long-term performance in a repository environment, but other factors also influence waste form performance. These include waste form geometry; density, porosity, and cracking; the presence of cladding; in-package chemistry feedback; etc. The paper proposes a formal definition of waste form "durability" which accounts for these effects. Examples from simple systems as well as from complex models used in the Total System Performance Assessment of Yucca Mountain are provided. The application of "durability" in the selection of bounding models is also discussed.

INTRODUCTION

In evaluating the performance of the proposed repository at Yucca Mountain, radionuclide release models are used in the Total System Performance Assessment (TSPA) to represent the long-term behavior of commercial spent nuclear fuel (CSNF), defense high-level waste glass (DHLW), and DOE spent nuclear fuel (DSNF) in the repository environment. However, each of these three models represents many types of waste. In particular, there are over 250 distinct types of DOE spent nuclear fuel that may be disposed in the potential repository.[1] Each of these spent nuclear fuel types has been categorized into eleven groups. A conservative model for the Hanford N Reactor fuel is the recommended surrogate to bound the performance of groups 2 through 11. (The exception, group 1, is naval spent nuclear fuel.) In addition, the inventory from the immobilized plutonium ceramic is averaged with the high-level waste glass inventory, and the higher intrinsic degradation rates for glass (in terms of mass released per unit area per unit time) are used to bound release.

To accurately compare the behavior of two dissimilar waste forms in a repository environment, a relative measure of waste form performance is needed. It is not sufficient to observe degradation rates alone when determining if a model for one waste type bounds release for another waste type. Other factors that influence radionuclide release rates include waste form geometry, density, cracking, porosity, and the presence of cladding. In a repository environment, additional factors such as in-package chemistry also play a role.

This paper proposes a simple definition of waste form "durability" that accounts for all the factors that influence radionuclide release and demonstrates that using the degradation model of a less durable waste form provides a conservative upper bound on release from a more durable waste form. Durability, therefore, is a measure of how well a given waste form retains its radionuclide inventory.

WASTE FORM DURABILITY

To define durability, start with an ideal system with a known intrinsic degradation rate, $k(t)$, and surface area, $A(t)$, where $k(t)$ has units of mass released per unit area per unit time. The rate at which mass, M, is released from the system is

$$\frac{dM}{dt} = A(t)k(t) . \qquad (1)$$

In the case where A and k are constant with respect to time, the time it takes the system to release all its mass is

$$T = \frac{M_0}{A_0 k} , \qquad (2)$$

where M_0 and A_0 are the initial mass and area and T could be defined as the total "lifetime" of the system. A

system with a long lifetime is said to be more "durable" than a system with a short lifetime.

Simplified Durability

The total lifetime of a system defined by Eq. (2) is unrealistic in that it assumes both A and k are constant. At time $t > 0$, there is less mass remaining in the system, and one might assume that surface area is decreasing with time while the degradation rate is dependent on environmental conditions such as temperature and pH. To account for this, we define durability as the expected lifetime of a system at time t:

$$D(t) = \frac{M(t)}{A(t)k(t)}. \qquad (3)$$

In this sense, durability can be thought of as the remaining lifetime of the system if all parameters (except mass) were held constant from time t onward.

Durability as defined by Eq. (3) accounts for more than the intrinsic degradation rate, $k(t)$. If the system undergoes cracking, as in DHLW glass, the effect is accommodated with a cracking factor included in $A(t)$. Note that cracking decreases durability by increasing the area. Similarly, porosity and density are included implicitly in the ratio $M(t)/A(t)$. For example, given two systems with the same intrinsic degradation rate and geometry but different densities, the one with the higher density will have a higher durability because there is a smaller area per unit mass.

Durability can be readily calculated as a function of time for simple systems such as spheres, cubes, and square cylinders. (A square cylinder has a diameter equal to its length.) If these shapes are assumed to degrade uniformly, then the geometry of each of these shapes can be characterized by a single, time-dependent parameter. When the intrinsic degradation rate is constant, this greatly simplifies Eq. (3) since it can be shown that

$$A(t) = A_0(1 - t/T)^2$$

and

$$M(t) = M_0(1 - t/T)^3$$

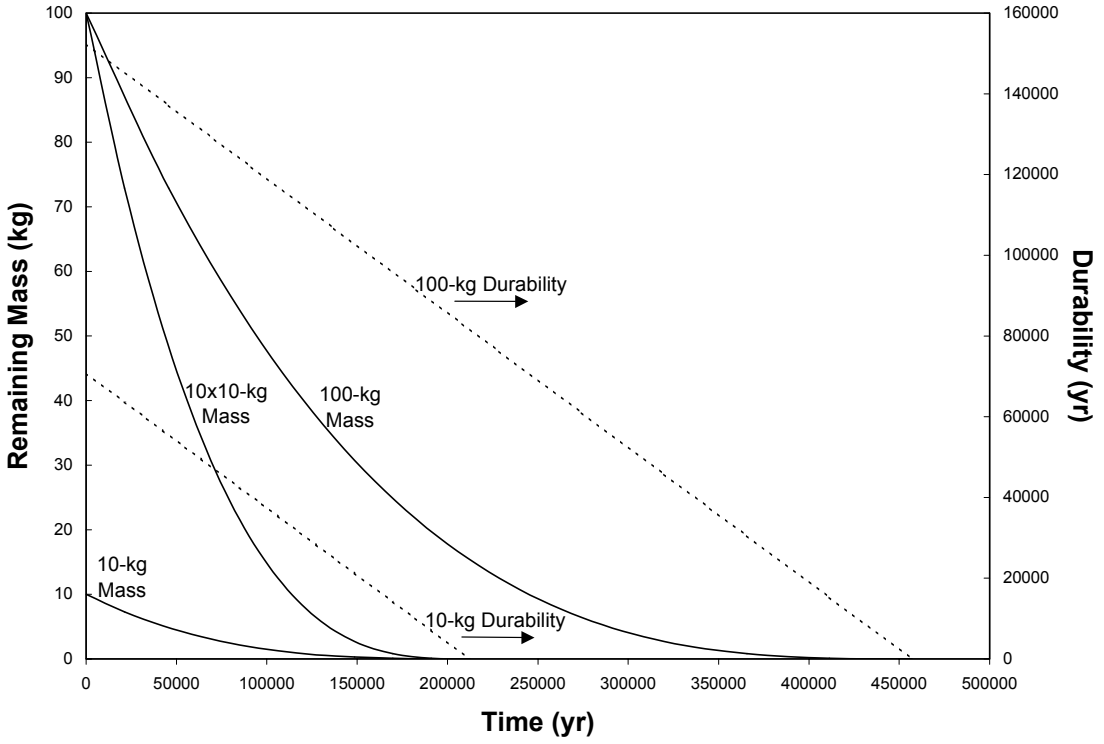

Figure 1: Remaining Mass and Durability as a Function of Time for 10-kg and 100-kg Spherical Waste Forms. Both waste forms are assumed to have the same degradation rate (1 g/m^2/yr) and the same density (2 g/cm^3). The remaining mass for a collection of 10, 10-kg waste forms is also shown.

Here, T is the total lifetime, which depends on geometry, density, and degradation rate. For single parameter systems with constant intrinsic degradation rate, it can be shown that the total lifetime is $T = \rho R_0/k$, where ρ is the density and $R_0 = 3V_0/A_0$ is the initial characteristic radius of the single parameter system.

To illustrate durability for a simple system, durability as a function of time for two different size spheres is plotted in Figure 1. The two spheres have the same density (2 g/cm^3) and constant degradation rate (1 g/m^2/yr) but different masses. For these systems, Fig. 1 shows that durability decreases linearly with time. This is because the ratio M/A decreases linearly with time.

The initial durability of the 100-kg sphere is over twice that of the 10-kg sphere. Furthermore, ten 10-kg spheres are no more durable than a single 10-kg sphere. The total lifetime of the 10-kg sphere is 212,000 years, while for the 100-kg sphere it is 457,000 years. Despite these long lifetimes, the majority of the mass is released much earlier: the 100-kg sphere releases 90% of its mass by approximately 250,000 years. Rather than total lifetime, it would be better to determine the mean time $\langle T \rangle$ for mass to be released from the sphere. This is defined as

$$\langle T \rangle = \frac{\int t dM}{\int dM}$$

The 10-kg sphere has a mean release time $\langle T \rangle$ of only 53,000 years, while the 100-kg sphere has a mean release time of 114,000 years.

For simple systems it can be shown that $\langle T \rangle$ is related to the initial durability by $\langle T \rangle = 3/4\ D(0)$. Therefore, initial durability gives an accurate measure of how well the two spheres would retain radionuclides. The conclusion from this simple example is that a single 100-kg waste form is preferred over 10 smaller 10-kg waste forms. This conclusion is not so obvious for more complicated systems.

Generalized Durability

The simple definition of durability presented above does not account for the presence of cladding or other features of a waste form that prevent or facilitate the release of radionuclides. These features can be accommodated by combining Eq. (3) with Eq. (1) to generalize the definition of durability as

$$D(t) = \frac{M(t)}{dM/dt}. \quad (4)$$

The rate at which mass is released from a system might depend on surface area, cracking, porosity, intrinsic dissolution rate, the presence of cladding, etc. Therefore, generalized durability is implicitly dependent on these factors as well.

Using the generalized definition of waste form durability presented in Eq. (4), it is possible to compare the performance of two dissimilar waste forms. For example, we can compare the performance of CSNF to DHLW glass, even though the two waste forms have distinctly different geometries, different radionuclide inventories, and degrade by different mechanisms. In addition, CSNF has the benefit of cladding, while DHLW glass suffers from cracking. Using source term degradation models used in the *Yucca Mountain Science and Engineering Report*,[1] (model SR00_042nm6) we have plotted the durability of CSNF (with and without cladding), DHLW glass, and DOE SNF at time of waste package failure in Fig. 2 for waste package failure times out to one million years. The values plotted represent the mean result from 300 realizations, calculated as

$$D = \left(\frac{1}{N} \sum_{i=1}^{N} D_i^{-1} \right)^{-1}$$

where D_i is the durability calculated for realization i and $N = 300$.

Because durability was calculated at the time of waste package failure, the mass and surface area of each waste form is at its known, initial value. Therefore, variations in durability as a function of waste package failure times are controlled by variations in the intrinsic degradation rate. (The absolute magnitude of durability still includes contributions from surface area, density, cladding, etc.) The general trend of durability in Fig. 2 is to increase with time This is because the repository temperature is decreasing, with a corresponding decrease in the intrinsic degradation rates for most waste forms. The exception to this is DOE SNF, which is represented by a degradation rate that is constant with respect to temperature.

The difference between clad and unclad CSNF durability reflects the difference in the initial fraction of failed fuel pins. In the CSNF source term model from the TSPA, the initial fraction of failed pins is approximately 8% (represented as a distribution in the

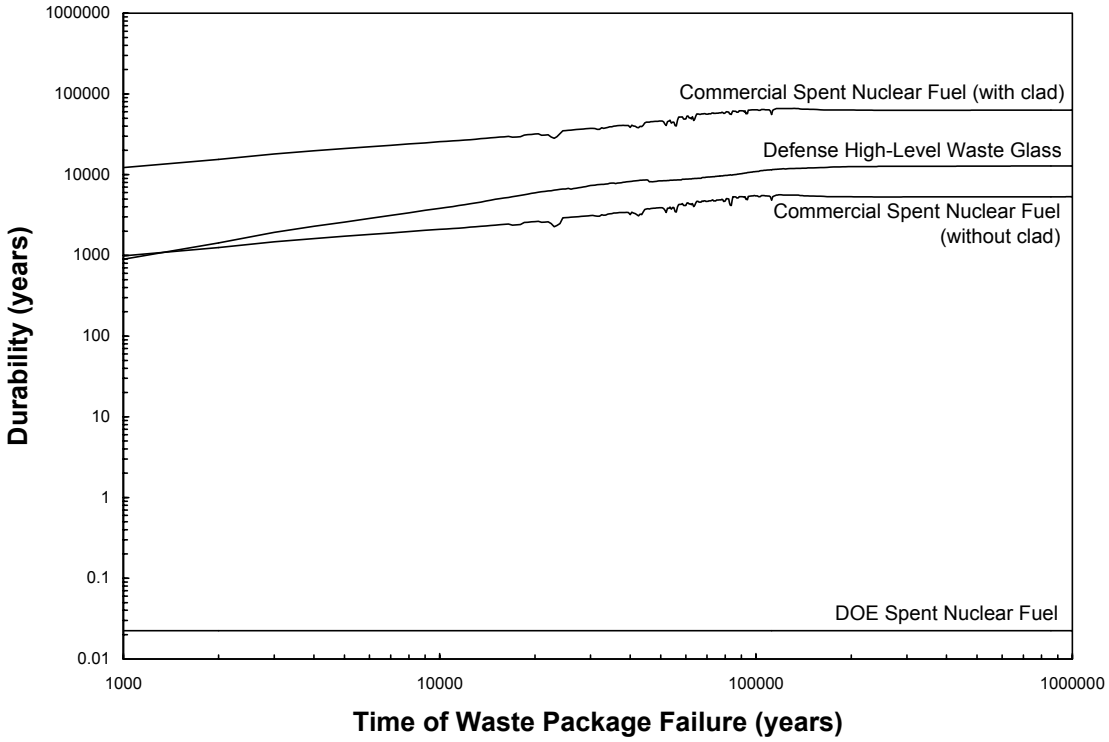

Figure 2: Initial Durability at Time of Waste Package Failure for Commercial Spent Nuclear Fuel (with and without cladding), Defense High-Level Waste Glass, and DOE Spent Nuclear Fuel.

model). In the unclad CSNF model, it is assumed that 100% of the fuel pins are failed at the time of waste package failure. This represents a 12-fold increase in the failure fraction, with a corresponding decrease in the durability.

Figure 2 only shows the *initial* durability at the time a waste package fails; it does not indicate how durability of a given waste form will change with time *after* a waste package fails. Based on the results of the simple systems shown in Fig. 1, we would expect that the actual durability of CSNF without clad, DHLW glass, and DOE SNF would decrease with time once a waste package fails. However, the models used in the TSPA assume that the specific surface area (A/M) of each waste form is constant; therefore durability will never decline toward zero as with the simple systems. The implication is that the waste forms will never completely degrade. In this regard, it is unlikely that the models in the TSPA are conservative for all times with respect to radionuclide release rates.

For clad CSNF, the cladding failure rate decreases with time. Initially, durability is controlled by the intrinsic degradation rate (combined with surface area, density, etc. of the fuel matrix). As the cladding failure rate decreases, it begins to control release, and durability begins to increase. The cladding failure rate approaches zero in the long term, therefore durability will tend towards infinity. Because of this, it is unlikely that the CSNF model is conservative for long-term modeling.

APPLICATION OF DURABILITY

Given the durability of two different waste forms, an appropriate bounding model for both waste forms can be obtained by using the model from the least durable waste form. This can be seen by rearranging Eq. (4) to read $dM/dt = M/D$. Total release from both waste forms is then bound by the expression

$$\frac{dM}{dt} \le \frac{M_1(t) + M_2(t)}{\min\left(D_1(t), D_2(t)\right)}. \qquad (5)$$

It is important to note that the identity of the least durable waste form could change with time, and it need not be the waste form with the highest intrinsic degradation rate, k. Hereafter, the time dependence on mass, M, and durability, D, is implicitly assumed.

When conducting a total system performance assessment (TSPA) of the proposed repository at Yucca Mountain, it is impractical to represent each of the hundreds of types of waste with a unique model. Instead, the various types of waste are grouped together and represented by a small group of models. To determine which model is appropriate for a given type of waste, it is necessary to compare the durability as defined by the model with the durability of the waste form in question.

In the TSPA, the total release rate from all the waste types i which are represented by model m can be written as

$$\frac{dM}{dt} = \frac{\sum_{i \in m} M_i}{D_m}, \tag{6}$$

where D_m represents the durability as defined by the model. The release rate predicted by the model bounds the total release of all the waste forms represented by the model if

$$\sum_{i \in m} \frac{M_i}{D_i} \le \frac{\sum_{i \in m} M_i}{D_m} \tag{7}$$

Equation (7) suggests that the reciprocal of the weighted average of the inverse durability of the waste forms represented by the model must be greater than the durability defined by the model:

$$D_m \le \left(\frac{\sum_{i \in m} M_i D_i^{-1}}{\sum_{i \in m} M_i} \right)^{-1} \tag{8}$$

A much simpler approach is to recognize that the release rate predicted by the model is guaranteed to bound the total release of all waste forms represented by the model if it can be shown for all times and for all $i \in m$ that

$$D_m \le D_i \tag{9}$$

The application of Eq. (9) may exclude waste forms from group m that might otherwise be included in the group by Eq. (8). However, for each new waste form that is a candidate for inclusion in a group, Eq. (8) must be reevaluated for all waste forms in that group.

SUMMARY

The definition of waste form durability presented here is a meaningful way to compare the performance of two dissimilar waste forms as it includes all the factors relevant to waste form performance. Although the definition of durability is straightforward, evaluating waste form durability as a function of time can be quite difficult, especially considering the complex interactions present in a repository environment. Nevertheless, it is possible to evaluate durability as defined by the repository models. If the durability of a waste form can be shown to be greater than the durability as defined by a model, then the model is said to bound the waste form. The comparison can be made regardless of the mechanisms of release, differences in waste form geometry, presence of cladding, or other factors that affect release rates.

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