

DEVELOPMENT OF A WASTE FORM PROCESS MODEL IN PFLOTRAN

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An important feature required in all geological disposal system modeling is proper representation of waste package degradation and waste form dissolution. These processes are often treated as batch operations, meaning they are zero-dimensional. However, waste package canister degradation or waste form dissolution are affected by near-field conditions, and thus they must be coupled to the computational domain through the exchange of information on local conditions. Accurate waste package and waste form degradation behavior is essential because processes occurring at the batch level also affect far field conditions through heat and mass transport by advection or diffusion. Presented here is the development and performance of the Waste Form Process Model, an integrated module for waste package canister degradation and waste form dissolution developed by Sandia National Laboratories within PFLOTRAN. PFLOTRAN is an open source, massively parallel subsurface simulator for multiphase, multicomponent, and multiscale reactive flow and transport processes in porous media. PFLOTRAN is used to model geologic disposal systems for the Spent Fuel and Waste Science and Technology (SFWST) Campaign under the Spent Fuel and Waste Disposition Program of the U.S. Department of Energy (DOE) Office of Nuclear Energy.

I. INTRODUCTION & BACKGROUND

The Spent Fuel and Waste Science and Technology (SFWST) Campaign, under the Spent Fuel and Waste Disposition Program of the U.S. Department of Energy (DOE) Office of Nuclear Energy, conducts research and development on geologic disposal of spent nuclear fuel and high-level nuclear waste. As part of this program, the Generic Disposal Systems Analysis (GDSA) work package [7] is responsible for developing a disposal system modeling and analysis capability that can evaluate disposal system performance for nuclear waste in geologic media, and can evaluate priorities in disposal research and development. The GDSA work package considers a wide range of disposal options, including salt, argillite, and crystalline host media, as well as deep borehole concepts.

Common to all geological disposal system models, regardless of the geologic media or repository design, are the waste packages containing nuclear waste. For any geologic disposal system model, proper representation of waste package degradation and waste form dissolution is essential. The GDSA work package is responsible for developing disposal system modeling and analysis capability that: (i) spans a wide range of disposal options, and (ii) must accurately consider unique waste package designs specific to each disposal system. Therefore, an ability to represent the large amount of possible waste package designs in a single simulator becomes a challenging problem. Modularity in the simulator design becomes vital to represent the large variety of waste packages in a manner that minimizes code maintenance and maximizes computational efficiency.

Waste package degradation and waste form dissolution are often treated as batch operations in disposal system simulations, meaning they are zero-dimensional. In reality, these processes are tightly coupled with near-field conditions, thus waste package degradation and waste form dissolution must be coupled to the computational domain, at minimum through the exchange of information on local conditions. More advanced representation of waste package degradation and waste form dissolution can be done using a multi-continuum technique, where waste packages occupy a 4th dimension in grid space that is physically coupled to the computational domain, rather than a zero-dimensional batch operation (see Section IV).

Presented here is the development and performance of the Waste Form Process Model, an integrated module for waste package degradation and waste form dissolution developed by Sandia National Laboratories within PFLOTRAN. PFLOTRAN is an open source, massively parallel subsurface simulator for multiphase, multicomponent, and multiscale reactive flow and transport processes in porous media [5]. PFLOTRAN is used to model geologic disposal systems for the evaluation of disposal system performance by the GDSA work package, as well as others under the SFWST Campaign [6; 7; 2; 9].

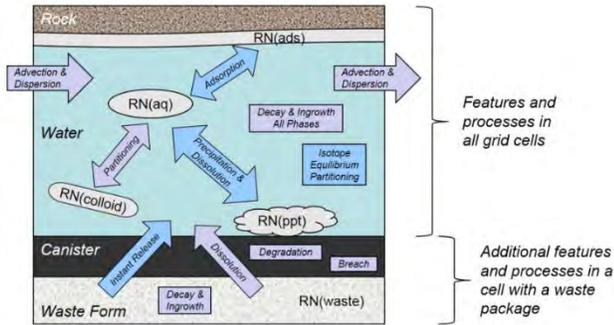


Figure 1. Schematic diagram of important processes affecting radionuclide (RN) concentration and transport, including the Waste Form Process Model.

II. THE WASTE FORM PROCESS MODEL FRAMEWORK AND WORKFLOW

The Waste Form Process Model is an integrated module that controls waste package degradation and subsequent waste form dissolution and radionuclide release to the geologic host media in a PFLOTRAN simulation. The evolution of all waste packages in a repository simulation is controlled by the Waste Form Process Model.

Workflow begins by instantiation of the process model within the PFLOTRAN input file, where several details of each waste package are defined (such as the location, radionuclide inventory, size, material, etc.). Once the simulation begins, each waste package evolves independently (e.g., waste package degradation, exposure of the waste form to the geologic host media (breach), dissolution of the waste form, and release of radionuclides). The Waste Form Process Model acts as a radionuclide source term to the rest of the PFLOTRAN simulation, with loose coupling to the local environmental conditions needed that affect waste package degradation or release rates (e.g., temperature, chemistry). Figure 1 illustrates the features and processes of waste package cells and the surrounding environment.

The process model is designed in a modular fashion, so that waste packages can be built from initially generic and standardized components. The components are customized to describe specific waste package or waste form inventories, properties, behavior, etc. A pre-defined set of waste package and waste form types are available for the user to choose from for common waste package designs and waste form types. Section III describes each component in detail.

Because PFLOTRAN is open-source, advanced users with unique applications are invited to create new waste package and waste form types within the standardized module framework by adding new source code. This can be achieved by simply using the existing module

components as a template, and has successfully been done for glass degradation at PNNL [8], for example.

Documentation for the Waste Form Process Model, as well as the rest of PFLOTRAN, can be found at http://www.documentation.pflotran.org/user_guide/cards/process_model_cards/waste_form_general_card.html.

III. THE WASTE FORM PROCESS MODEL COMPONENTS

The new Waste Form Process Model in PFLOTRAN consists of three main components: (a) the waste form canister, (b) the waste form object, and (c) the waste form release mechanism (see Figure 2).

III.A. The Waste Form Canister

The waste form canister conceptual model addresses (1) the timing of canister breach and (2) the performance of the canister after breach. In this conceptual model, the status of the canister is defined by two abstract terms, *canister vitality* and *canister performance*. Canister vitality is a normalized measure of remaining time or remaining canister wall thickness before canister breach, and canister performance is a normalized measure of the physical ability of the canister to contain the source. Initially, both terms have a value of 1. Before canister breach, while corrosion reduces the time remaining or canister wall thickness remaining before canister breach, the canister vitality decreases. When it reaches zero, the canister is breached and canister performance begins to decrease. See Section 3.2.4 of [6], for details.

To date, canister performance has not yet been implemented in the waste form canister component, but a framework for canister vitality is complete. In this framework, canister vitality is initialized to 1, and is reduced at each time step by the effective canister vitality degradation rate R_{eff} , according to

$$R_{eff} = R e^{C \left(\frac{1}{333.15} - \frac{1}{T(LX)} \right)} \quad (1)$$

where R is the base canister vitality degradation rate at 60°C, T is the local temperature (in Kelvin), and C is the canister material constant. This equation assumes that reaction rates are a function of temperature as described by the Arrhenius equation. For general corrosion, R would represent the general corrosion rate at 60°C in units of L/T, and the associated canister vitality would be a measure of the remaining normalized canister thickness before breach.

The canister vitality degradation rate as it has been initially implemented this year provides a framework upon which more mechanistic processes can interface. Coupling

and interfacing the canister degradation model with more mechanistic processes (such as general corrosion, stress corrosion cracking, pitting corrosion, microbiologically-influenced corrosion, rock fall, etc.) is planned to start in FY17.

Once canister vitality drops below zero, the canister is considered breached, and a Boolean flag is turned on for the waste form object inside of it. The user may alternatively specify the canister breach time for each waste package. This functionality was included to allow for early breach times, or to guarantee a breach time if the available options for degradation rate are insufficient.

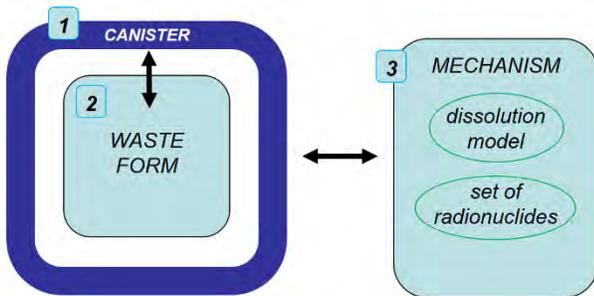


Figure 2. The three main components of PFLOTRAN's Waste Form Process Model consists of the waste form canister, the waste form object, and the waste form mechanism.

III.B. The Waste Form Object

The second component of the Waste Form Process Model is the waste form object. This object is generic and contains only the information that is required by all waste form types. The user defines each waste form object's location in the domain, as well as its initial volume, and exposure factor (a surface area multiplying factor to the waste form's effective dissolution rate). Within the waste form object, the value of its effective dissolution rate is stored. Each waste form object has a pointer to the waste form mechanism (the third component of the process model) that describes waste form type-specific information. The dissolution equation that defines the effective dissolution rate is obtained from the waste form mechanism. The waste form object also stores the concentrations of the radionuclide inventory. The initial radionuclide inventory is obtained from the waste form mechanism.

Radionuclide decay and ingrowth is internally calculated for the set of radionuclides in each waste form according to a 3-generation analytical solution derived for multiple parents and grandparents and non-zero initial daughter concentrations (see Section 3.2.3 [7]). The solution for radionuclide concentration within the waste form is obtained explicitly in time. Internal calculation of radionuclide decay and ingrowth also allows the

calculation of instantaneous release fractions for certain radionuclides upon canister breach. A fully-implicit solution for multiple-parent, multiple-daughter radioactive decay and ingrowth is planned.

Upon canister breach, the waste form object begins to dissolve according to the dissolution model that is defined by the waste form mechanism to which the waste form object points. Waste form volume decreases accordingly. The effective dissolution rate along with the radionuclide concentrations in the waste form, determines the source term (radionuclide release rate) for each waste form.

III.C. The Waste Form Mechanism

The third component of the Waste Form Process Model is the waste form mechanism. In contrast to the other two components, this object is specific to the type of waste form being simulated and contains information which defines the behavior of each specific waste form type. The mechanism contains the value of the waste form bulk density, the set of initial radionuclides (initial mass fractions, molecular weights, decay rates, daughter species, and instantaneous release fractions), and a pointer to the waste form dissolution model. In some cases, it also stores the waste form specific surface area.

Since a performance analysis simulation typically contains hundreds or thousands of waste form objects but only a few waste form "types," separating the waste form type-specific information into the waste form mechanism improves modularity and numerical efficiency. An additional benefit of the modularity is that new waste form types can easily be created in PFLOTRAN by simply creating new waste form mechanisms. Moreover, coupling to external dissolution models, such as FMDM [3], is easily accomplished through the modularity provided with the waste form mechanism.

Currently, four types of waste form mechanisms have been implemented. Details of each mechanism are described below. For three of the waste form mechanisms, a series of three figures are included to demonstrate the capability of the Waste Form Process Model. The figures portray the evolution of a single waste form inside a cube of 27 (3×3×3) 1-m³ grid cells. The simulation assumes no fluid flow, no diffusive flux across the domain boundaries, and a constant temperature of 25°C. The simulations were run for 100 million years (10⁸ years). Each are identical except for the waste form mechanism used. A schematic of the computational domain is shown in Figure 3.

- no fluid flow
- no diffusive flux across boundaries
- $3 \times 3 \times 3 = 27$ grid cells
- 1 m^3 grid cells

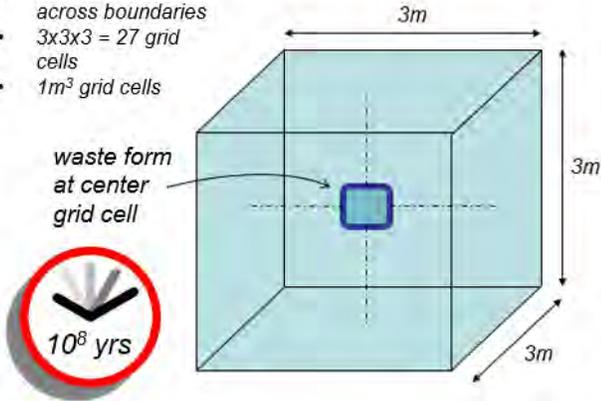


Figure 3. Set up of the computation domain for the Waste Form Process Model demonstrations.

III.C.1. The GLASS Mechanism

High level waste in the form of glass logs are simulated using the GLASS mechanism. The glass dissolution model used in this mechanism is according to Kienzler *et al.* (2012),

$$r(T) = 560e^{\frac{-7397}{T(t,x)}} \quad (2)$$

where $r(T)$ is the effective dissolution rate ($\text{kg-glass m}^{-2} \text{ day}^{-1}$) and T is the temperature (Kelvin) at the current time (t) and location (x) of the waste form. The effective dissolution rate is converted to a fractional dissolution rate by multiplying $r(T)$ by the specific surface area (in units of L^2/M), which is provided by the user. Figure 4, Figure 5, and Figure 6 demonstrate a Savannah River Glass waste form using this mechanism. The initial inventory was chosen based on year 2038 [1; 9].

III.C.2. The Instantaneous Mechanism

For the Instantaneous mechanism (currently called DSNF mechanism in PFLOTRAN), at the time step when canister breach occurs the entire radionuclide inventory of the waste form is released over the length of the time step. Concurrently, the volume of the waste form is reduced to zero. Metallic defense-related spent nuclear fuel (DSNF) is simulated using this mechanism. Figure 7, Figure 8, and Figure 9 demonstrate a waste form composed of defense-related spent nuclear fuel in the 300W – 500W bin [10; 9].

III.C.3. The FMDM Mechanism

Used nuclear fuel (composed of uranium dioxide) is simulated using the Fuel Matrix Degradation Model (FMDM) mechanism. This mechanism also demonstrates how external dissolution models can be coupled to PFLOTRAN. The dissolution model used is obtained via coupling to the FMDM by calling a single external subroutine developed by [3]. Details regarding the FMDM conceptual model and algorithmic design are provided by [3]. While PFLOTRAN-FMDM coupling has been successfully established, it is not yet optimized. Optimization is needed to speed up FMDM simulation. FMDM run time has a major impact on the overall run time of a repository simulation when there are a large number of waste packages. Figure 10, Figure 11, and Figure 12 demonstrate a waste form composed of used nuclear fuel using the FMDM mechanism. Initial inventory of selected radionuclides is based on 30-year decay time, commercial PWR assemblies, 60,000 MWD/MTHM burn-up, and 4.73% enrichment [1]. We note that the dissolution rates are higher than expected, possibly due to numerical dispersion caused by insufficient grid resolution in the FMDM. This may be remediated by using a finer grid in FMDM.

III.C.4. The CUSTOM Mechanism

To allow additional flexibility, the CUSTOM mechanism was created so that a user can specify a fractional dissolution rate (in units of $1/T$), or a waste form dissolution rate that is based on specific surface area (in units of $\text{M}/\text{L}^2/T$). If the user specifies a surface area dependent dissolution rate, a specific surface area (in units of L^2/M) must also be provided.

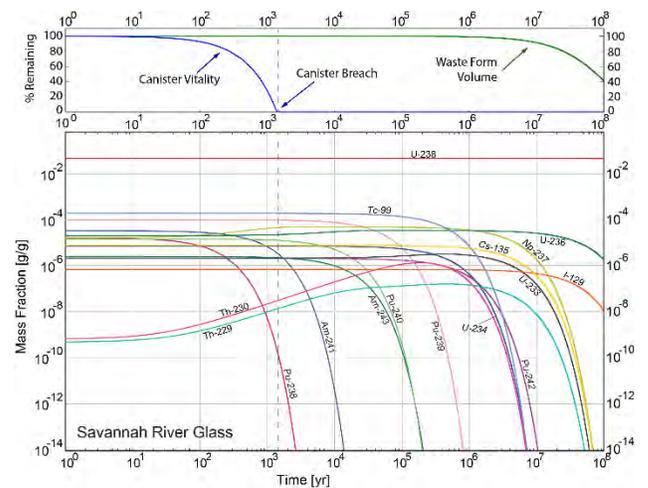


Figure 4 Canister vitality, waste form volume, and radionuclide mass fraction in a HLW Savannah River Glass waste form using the GLASS mechanism.

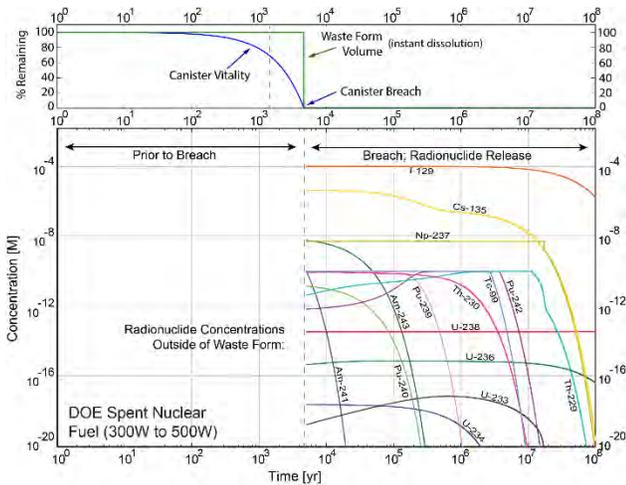


Figure 9 Canister vitality, waste form volume, and concentrations outside of a defense-related spent nuclear fuel (300W-500W bin) waste form using the Instantaneous mechanism.

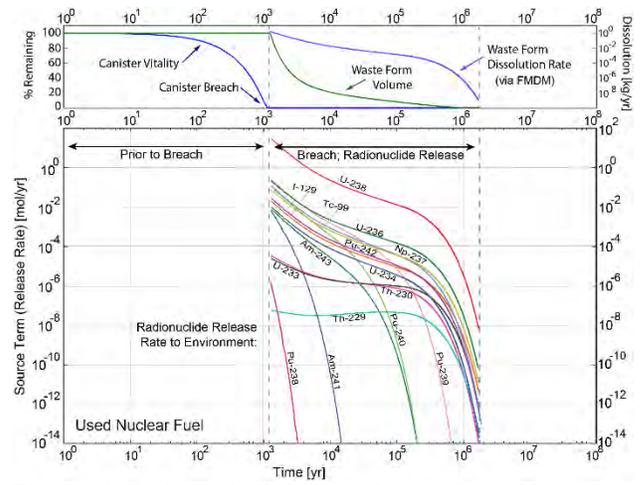


Figure 11 Canister vitality, waste form volume, waste form dissolution rate, and radionuclide release rate (source term) in a used nuclear fuel waste form using the FMDM mechanism.

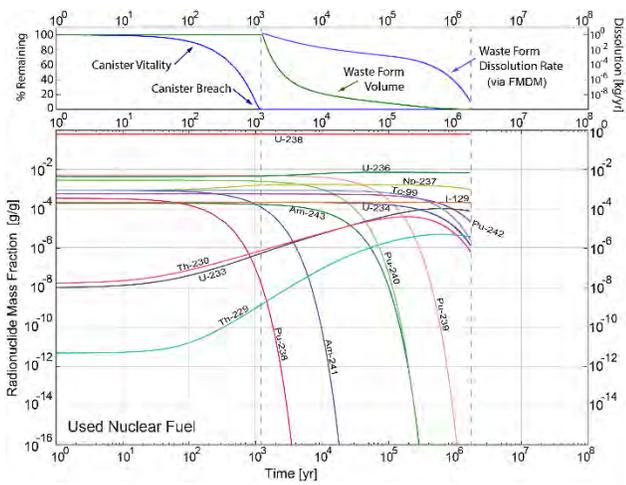
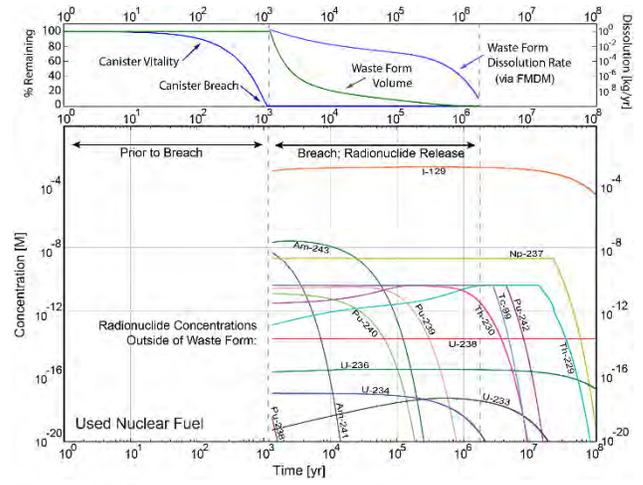


Figure 10 Canister vitality, waste form volume, waste form dissolution rate, and radionuclide mass fraction in a used nuclear fuel waste form using the FMDM mechanism.



IV. A MULTI-CONTINUUM APPROACH

More advanced representation of waste package degradation and waste form dissolution can be done using a multi-continuum technique, where waste packages occupy a 4th dimension in grid space that is physically coupled to the 3D computational domain, rather than a zero-dimensional batch operation. While a multi-continuum approach is more complex to set up numerically, it is more ideal for representing some sub-grid scale waste package and waste form processes without refining the main computation grid at each waste package cell. Thus, the multi-continuum technique can save computational expense.

The multi-continuum approach described here is proposed as future work, and will allow for increasingly mechanistic waste package and waste form processes to be implemented. For example, waste package designs are often composed of, or surrounded by, several material layers. Each layer serves a specific purpose in isolating the waste form inside from the host rock material. The discretization of the computational domain is often too coarse to explicitly resolve details of each waste package. Therefore, many processes, such as canister corrosion or radionuclide diffusion across a compromised material layer, are not resolved and must be parameterized.

By implementing a multi-continuum grid, the cells that host waste packages are given a 4th dimension. On this extra dimension, physical processes such as diffusion can be solved in 1D, and related back to the 3D grid. There is minimal growth in grid size because refinement does not occur on the 3D grid (which would increase the number of grid cells locally by order n^3); Rather, refinement occurs on the 4th dimension of the grid (which only increases the number of grid cells locally by order n). The multi-continuum approach is different from running a nested 1D model within the cells that host waste packages because the grid is continuously defined across all dimensions and all physics and chemistry are fully coupled.

V. CONCLUSIONS

Presented here is the development and performance of the Waste Form Process Model, an integrated module for waste package degradation and waste form dissolution developed by Sandia National Laboratories within PFLOTRAN, and used to model geologic, nuclear waste disposal systems for the evaluation of disposal system performance. Modularity in its design allows representation of a large variety of waste packages while minimizing code maintenance efforts and maximizing computational efficiency. Figures demonstrating the performance of several waste form types are presented.

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