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Sensitivity and Uncertainty Analysis of FMD Model Choice for a Generic Crystalline Repository

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SUMMARY

This paper applies sensitivity and uncertainty analysis to compare two model alternatives for fuel matrix degradation for performance assessment of a generic crystalline repository. The results show that this model choice has little effect on uncertainty in the peak ^{129}I concentration. The small impact of this choice is likely due to the higher importance of uncertainty in the instantaneous release fraction and differences in epistemic uncertainty between the alternatives.

INTRODUCTION

High-fidelity fuel degradation modeling is computationally intensive, so alternative models can provide tangible computational benefits, particularly for analyses where many simulations are required. One alternative for such modeling utilizes a fractional dissolution rate (FDR). Another modeling alternative utilizes machine learning to construct a surrogate model of the high-fidelity fuel matrix degradation (FMD) mechanistic model. Both provide computational efficiency compared to the high-fidelity mechanistic model which is intractable at a detailed repository scale.

This work examines how these two modeling approaches to waste form degradation affect performance predictions for a generic crystalline repository reference case. The surrogate model alternative, built from high-fidelity FMD training data, has more detailed predictions of the waste form behavior over time because it incorporates degradation rates changing with time. We examine how these FMD model alternatives influence the behavior of the system.

The United States Department of Energy (DOE) is developing a state-of-the-art simulation software toolkit, the Geologic Disposal Safety Assessment (GDSA) Framework, for probabilistic post-closure performance assessment (PA) of systems for deep geologic disposal of nuclear waste (https://pa.sandia.gov). The characterization and quantification of uncertainty (UQ) is an integral component of PA. Sensitivity analysis (SA) is used to identify uncertain parameters and processes that dominate uncertainty in performance metrics for performance assessment studies of geologic radioactive waste disposal sites.

In previous case studies, we performed SA studies on a generic crystalline reference case, first studying the effects of epistemic uncertain parameters [1] and then extending this to account for the spatial heterogeneity represented by a family of discrete fracture networks (DFNs) [2]. This work represents a further extension: adding the effect of a constitutive chemistry model for nuclear fuel dissolution. In this paper, we present a SA case study that seeks to separate the effect of epistemic parameters, spatial heterogeneity as represented by DFNs, and model choice as represented by the form of the FMD alternative model.

This paper summarizes the crystalline case and the two FMD model alternatives, presents the sensitivity and uncertainty analysis framework used to assess the influence of the FMD alternatives, and discusses how those effects compare to other uncertainties.

CRYSTALLINE REFERENCE CASE

This paper presents a generic reference case, referred to as the crystalline reference case, which is not representative of a specific site. The case is meant to be illustrative for the purpose of developing and demonstrating PA methods within the GDSA Framework. The crystalline reference case is a generic repository modeled in a crystalline rock formation. This type of repository would be placed in highly impermeable fractured rock, where flow occurs predominantly through the fractures. The repository model includes nuclear waste packages (WPs) which breach based on an uncertain normalized general corrosion rate. The WPs consist of a stainless steel canister and stainless steel overpack; they are expected to fail between ten thousand years and one million years post closure [3]. A key Quantity of Interest (QoI) is the maximum ^{129}I concentration in the aquifer over time, as well as its overall peak value (the maximum over all time points). Transport of ^{129}I is modeled from the WPs through the repository and surrounding host rock based on additional uncertain permeability, porosity, and radionuclide release parameters. The case is modeled in PFLOTRAN as a single-phase liquid flow and transport simulation.

Fig. 1 shows an example of the crystalline model. Concentrations of ^{129}I are monitored at pre-specified points in the model domain called observation points, and the maximum concentration is tracked throughout the aquifer.

Discrete Fracture Networks

The model domain is approximately 3000 m in length, 2000 m in width, and 1260 m in height [4]. The repository is located at a depth of 585 m. Forty-two disposal drifts contain 40 12-PWR WPs each (1680 total WPs). Drifts are backfilled with bentonite buffer and are surrounded by a 1.67-m thick disturbed rock zone. Each WP has a 5.225 metric tons of heavy metal (MTHM) inventory, so the total inventory for the crystalline reference case is 8,778 MTHM [5]. Within the repository, grid cells are as small as 1.67-m on a side; elsewhere grid cells are 15-m on a side. The model domain contains approximately 4.8 million grid cells. Additional information on the grid and dimensions may be found in a previous conference paper, available for download at https://pa.sandia.gov [4].

Fractured crystalline rock is modeled using stochastic discrete fracture networks, which are two- dimensional ellipses distributed in the three-dimensional model domain. The fracture networks are generated using dfnWorks [6], and mapped to the equivalent continuous porous medium domain using mapDFN.py, a code that approximates hydraulic fracture properties by calculating and assigning permeability and porosity on a cell-by-cell basis [4].

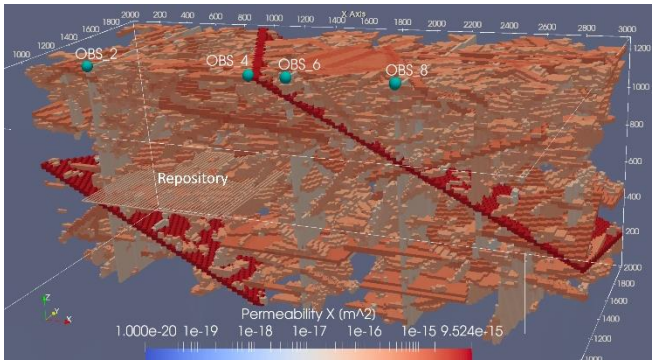

Fig. 1 Cut-away of a DFN realization mapped to porous medium grid colored by permeability, showing the full repository and far half of the model domain. The three-dimensional structures inside the domain are the repository, five deterministic deformation zones (two are colored red due to their high permeability, three subvertical zones are cream-colored), the fractures of a stochastically generated fracture network colored peach/orange throughout the model domain, and four teal observation points placed in the granite just below the aquifer (not pictured).

The discrete fracture networks are generated probabilistically over the domain, drawing from distributions of fracture characteristics such as fraction density, radius, orientation, and centroid location. Details about the DFN characterization and generation process can be found in [1, 2].

All DFNs are generated to have similar bulk properties, However, their generation introduces spatial heterogeneity; DFNs generated with the same distributions for fracture radii, orientations, and centroid locations are still significantly different from each other. The spatial heterogeneity between DFNs is a significant source of uncertainty affecting the QoIs, as discussed in the Results section.

FUEL MATRIX DEGRADATION ALERNATIVES

Fractional Dissolution Rate

The fractional dissolution rate model uses a fractional dissolution rate and the radionuclide concentrations in the waste form to determine the mass dissolution rate for each waste form [7, 8]. It is a fairly simple analytic model which is heavily dependent on the fractional dissolution rate of spent nuclear fuel, denoted as *rateUNF*.

Artificial Neural Network (ANN) surrogate

The Fuel Matrix Degradation model [9] is a complex chemistry model for calculating spent fuel degradation rates as a function of radiolysis, alteration layer growth, and diffusion reactants through alteration layer. It incorporates mixed potential and analytical radiolysis models. It must be called each time step for each WP, making it a very costly process model. To combat this cost, time-series FMD training data was obtained offline from the full process model and used to train an artificial neural network surrogate model alternative to the FMD model.

The ANN had 400k training points consisting of six inputs (fuel temp, dose rate, chemical species concentrations) and one output (UO_2 surface flux/fuel dissolution rate) [10, 11]. A two-layer ANN with 64 nodes per layer was utilized, resulting in 4673 parameters (weights and bias terms) that were estimated based on the training data. The surrogate ANN alternative to the FMD model was called from PFLOTRAN with a similar API as the full FMD model. Examples of prediction accuracy with the ANN surrogate are shown in Fig. 2. Note that the mean absolute error on the testing set was 8.26×10^{-4} mol/m^2/year and a relative test error of about 25%. This ANN has some error but matches the overall trend well.

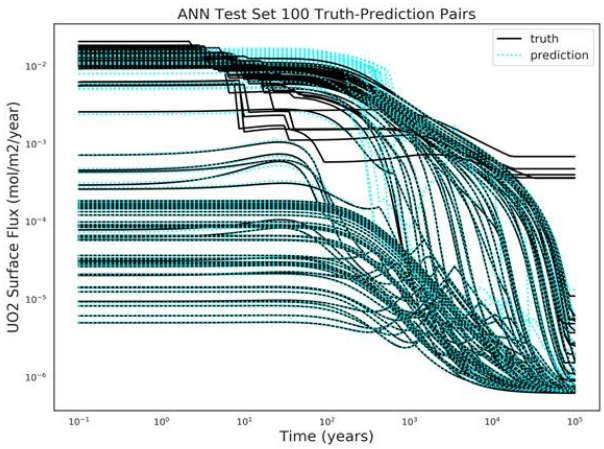

Fig. 2 Comparison of ANN surrogate to full process model results.

UNCERTAINTY AND SENSITIVITY ANALYSIS

SA for the crystalline reference case was performed using 1000 PFLOTRAN simulations for each FMD model alternative. The uncertainties for these 1000 simulations were sampled from a dual-loop structure, which separates spatial uncertainties from parametric uncertainties. The outer loop consists of 25 spatial realizations, which comprise uncertainty in the DFNs and the assignment of WP degradation rates between WPs. Forty simulations were performed for each of these spatial realizations using different samples for the glacial aquifer permeability (*kGlacial*), disturbed rock zone permeability (*permDRZ*), buffer porosity (*pBuffer*), buffer permeability (*permBuffer)*, instant release fraction (*IRF*), and WP normalized corrosion rate parameters (*meanWPrate, stdWPrate*).

The same 1000 samples were used in the PFLOTRAN simulations for each of the FMD model alternatives with one exception. Simulations with the FDR model included an additional uncertain parameter, the fractional dissolution rate (*rateUNF*). Hence, simulation-to-simulation differences demonstrate the extent to which the alternative model choice affects performance metrics.

Global SA was performed on each set of 1000 simulations using a second order polynomial chaos expansion (PCE) surrogate to estimate main and total effect Sobol' indices with Dakota [12]. The main effect index describes the proportion of the variance in the QoI that the PCE surrogate attributes to a variable on its own. The total effect index describes the proportion of the variance in the QoI that the PCE attributes to a variable and its two-way interaction with other variables.

The DFNs present a challenge for SA because DFN uncertainty is not defined parametrically. Quantities summarizing important features of the DFNs were calculated and included as proxy variables in the SA so the PCE surrogate could account for some of the influence of DFN uncertainty. These quantities are the average number of intersections between fractures (AveDegree), the number of fractures intersecting the repository (Intersections), and the relative shortest travel time (STT) [1]. Because the DFNs are repeated samples (there are 40 epistemic simulations per DFN), SA results may be biased towards attributing variance to DFN summary quantities. Because of this, the SA is run twice, once without DFN quantities and once with them. We interpret the results together assuming the true sensitivity indices are between the estimated values for the two analyses.

RESULTS

The maximum ^{129}I concentration in the aquifer is plotted over time in Fig. 3 for simulations with the FDR model (black) and with the ANN surrogate (pink dashed). The concentrations are very similar between the simulations with the two alternatives to the FMD model, but some differences are apparent towards the end of the simulation, where there is more variability in concentrations for the FDR model simulations. The mean and standard deviation of the concentrations at 1 million years are 3.66×10^{-9} [M] and 6.74×10^{-9} [M] respectively for the FDR simulations and 3.41×10^{-9}[M] and 5.00×10^{-9} [M] respectively for the ANN simulations.

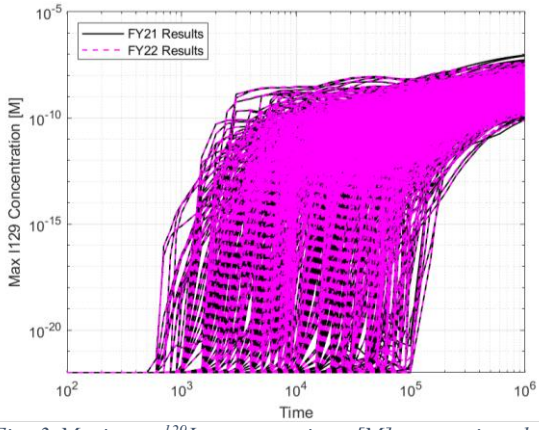

Fig. 3 Maximum ^{129}I concentrations [M] comparison between the simulations with the ANN and FDR model.

The difference in uncertainty between simulations for FMD model alternatives is also seen in peak ^{129}I concentration empirical cumulative distribution functions (ECDFs) plotted in Fig. 4. The grey curves are the ECDFs for each spatial realization; red/black curves show the ECDFs for all simulations with the ANN surrogate (red) and the FDR model (black). The domains for the FDR ECDFs are wider, indicating more variation in peak ^{129}I concentrations within a spatial realization when the FDR model is used.

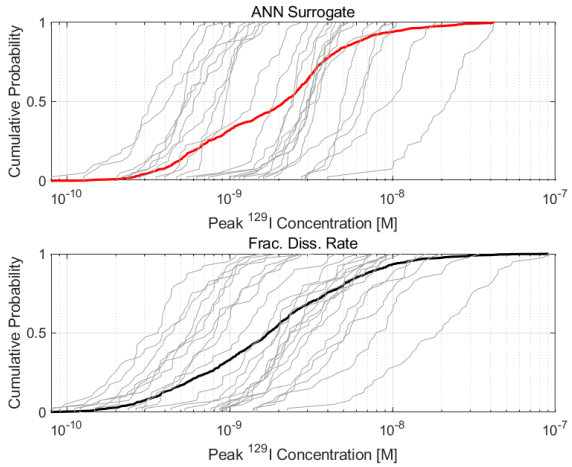

Fig. 4 Peak ^{129}I ECDF functions for each spatial realization (grey) and overall (black/red) for the ANN and FDR simulations.

The increased uncertainty in the FDR simulations is likely due to the incorporation of uncertainty in that model via the *rateUNF* parameter. No analogous uncertainty is currently included in the ANN surrogate model.

Sobol' indices are plotted for the ANN surrogate simulations in Fig. 5. The top results are for the analysis without the DFN quantities, and the bottom results are for the analysis with the DFN quantities. These results indicate that *kGlacial* and *IRF* are the dominant parametric uncertainties

driving variation in peak ^{129}I concentrations. The Intersections and AveDegree quantities are the dominant spatial uncertainties.

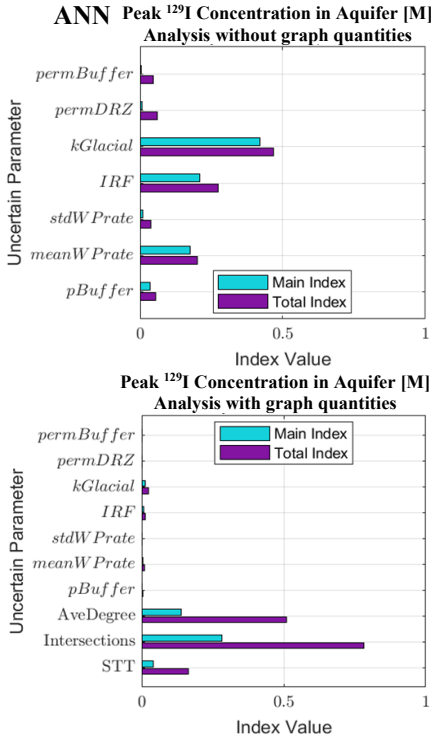

Fig. 5 Sobol' index estimates for peak ^{129}I concentrations for the simulations with the ANN model

These results can be compared to those for FDR simulations, plotted in Fig. 6. For these simulations, the dominant uncertainty becomes the FDR parameter uncertainty, *rateUNF,* with *kGlacial* and *IRF* having minor effects. The analysis with the DFN quantities identifies the same important spatial uncertainties as for the ANN simulations. Scatter plots of all of the input parameters versus the peak ^{129}I concentration support these SA results (see [13]).

The analysis for peak ^{129}I concentration was repeated for the maximum ^{129}I concentration at each time step. These results are plotted in Fig. 7. Only the results without the DFN quantities are included here because these results highlight the importance of the *rateUNF* uncertainty as well as the timing of importance for IRF and *rateUNF*. Results with the DFN quantities are in [13]. According to these results, the *IRF* parameter significantly contributes to the maximum ^{129}I concentration regardless of which FMD model is used. However, this parameter drops in importance towards the end of the simulation, where *kGlacial* becomes significant for the ANN simulations and *rateUNF* becomes significant for the FDR simulations.

This analysis provides an explanation for why there is so little difference between the ^{129}I concentrations for the ANN and FDR simulations in Fig. 3 until the end of simulation; the *IRF* dominates ^{129}I release early on, so the FMD model only begins to have a significant effect in the second half of the simulation time.

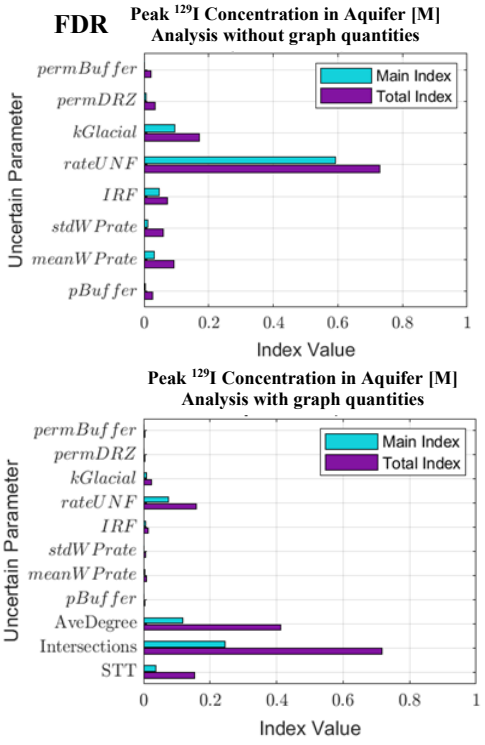

Fig. 6 Sobol' index estimates for peak ^{129}I concentrations for the simulations with the FDR model

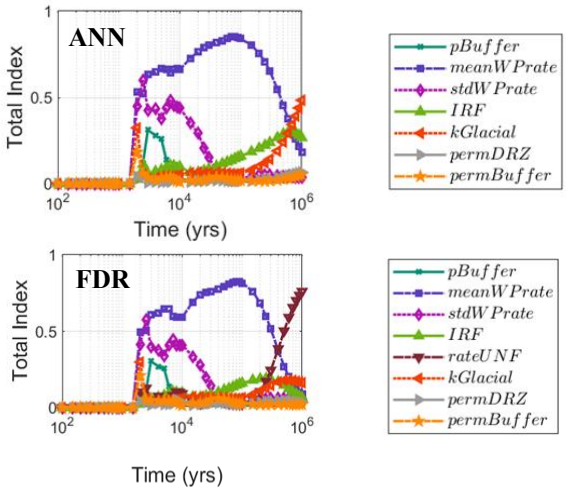

Fig. 7 Time-dependent SA results for the max ^{129}I concentration [M] for ANN simulations (top) and FDR simulations (bottom)

The uncertainty and SA results show that, overall, the choice for FMD model alternative only has a minor effect on ^{129}I concentrations. The timing for release of ^{129}I is similar between the two models, but the mean and variance of the peak ^{129}I concentration are slightly smaller at 1M years for

the ANN surrogate alternative. The similar high-level behavior of the models may be due to WP breach and instantaneous release dominating concentrations for much of the simulation time.

We expect the ANN model to provide a more detailed prediction of the waste form behavior over time because it incorporates dose rates changing with time and accounts for the chemistry in the local environment. Future work on the FMD model alternatives will include uncertainty in the environmental chemistry for the ANN model. This may affect the relative effects of the different FMD model alternatives, which could be assessed using a similar SA study.

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