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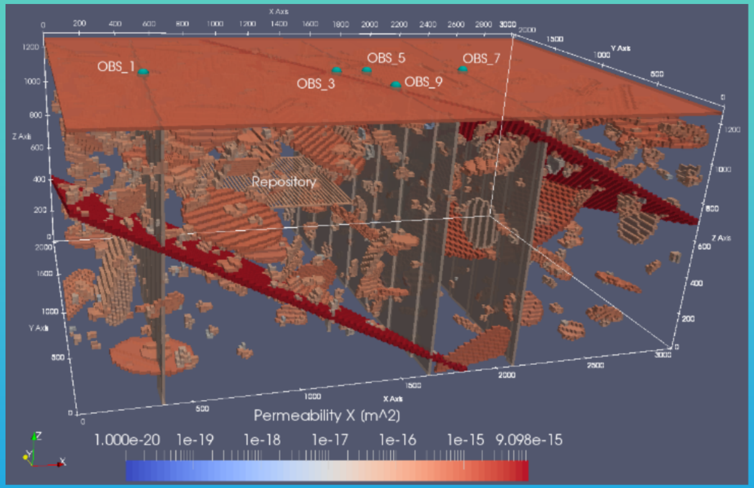
Spent Fuel and Waste Science and Technology (SFWST)

Clean. Reliable. Nuclear.



Neural-ODE Surrogates for Fuel Degradation Processes in Nuclear Waste Repository Simulations

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Sandia ML DL Workshop

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Acknowledgements and Disclaimers

- Acknowledgements:
  - Funding by the Department of Energy (DOE), Office of Nuclear Energy, Spent Fuel and Waste Science and Technology

- Disclaimer:

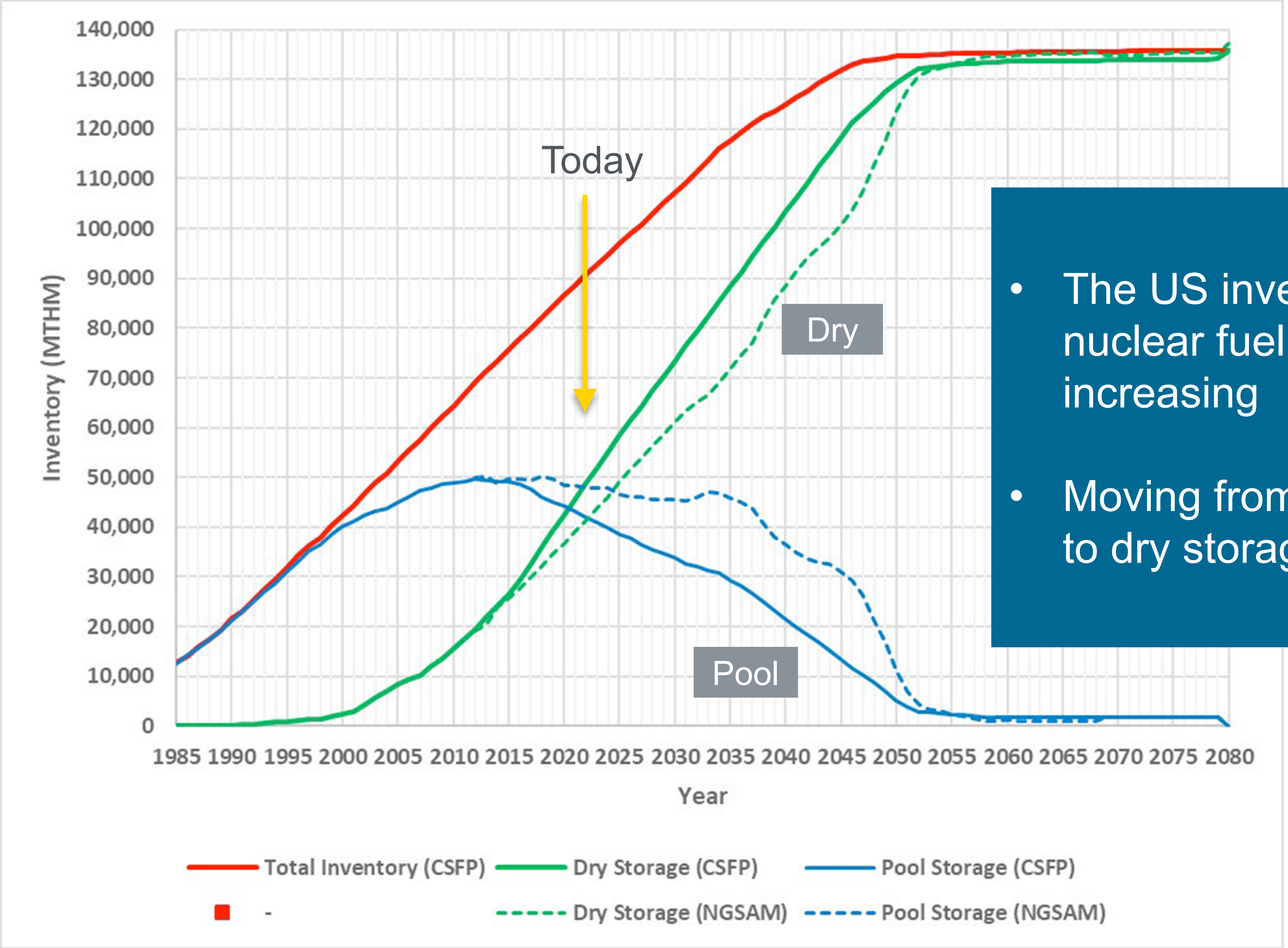
Overview

- Computational Modeling of Spent Nuclear Fuel
- Training Data
- Neural-ODE Surrogates
  - Methods
  - Logistic Decay Toy Problem
  - Application to Spent Nuclear Fuel
- Conclusions and Future Work

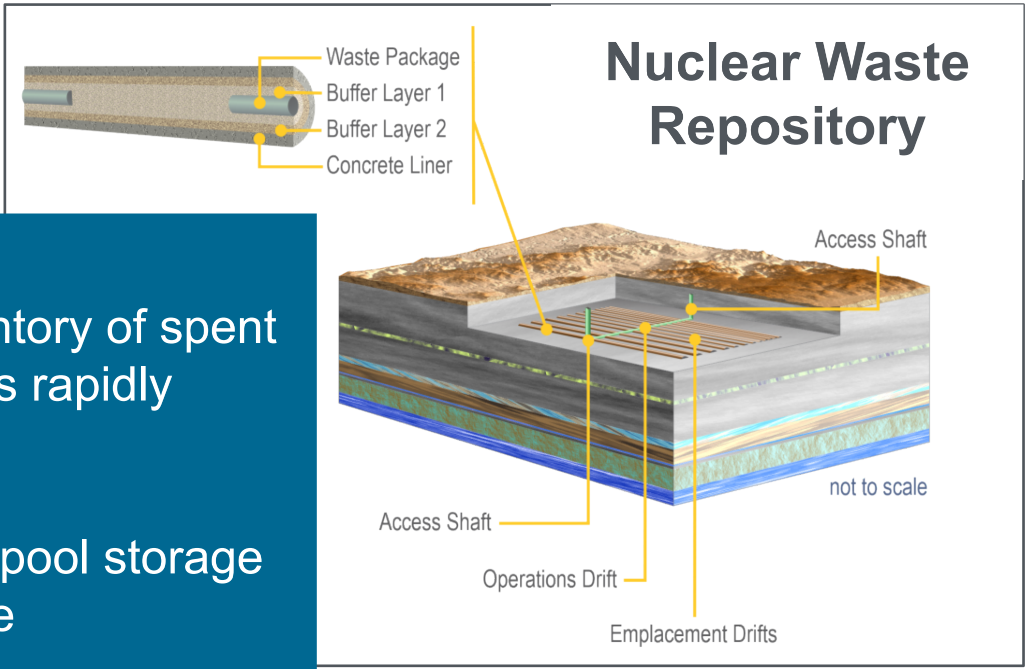
Computational Modeling of Spent Nuclear Fuel

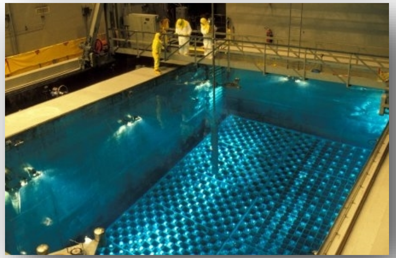
More Spent Nuclear Fuel in Dry Storage

Storage Projections (2 models)



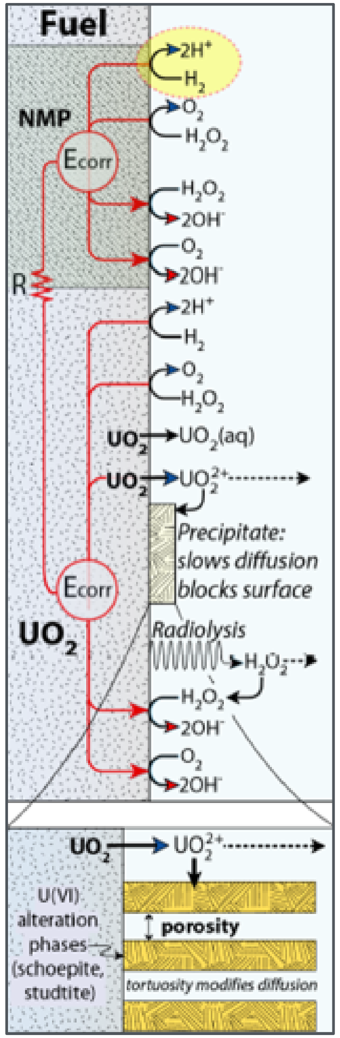
- The US inventory of spent nuclear fuel is rapidly increasing
- Moving from pool storage to dry storage



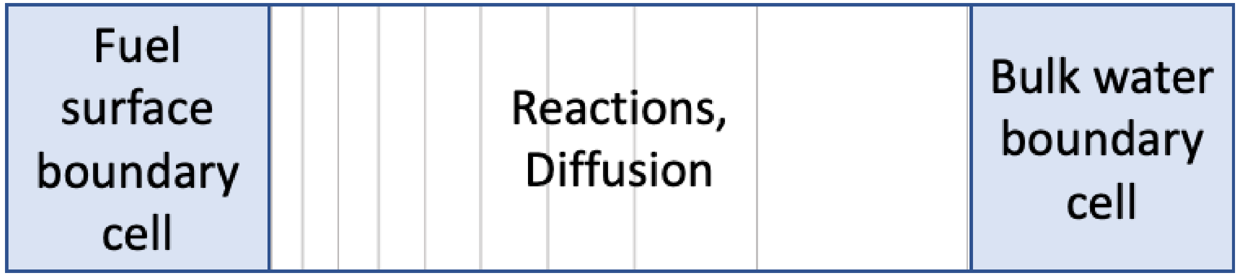


[1] Adapted from Freeze et al. (2021, Figure 2-3)

Fuel Matrix Degradation Model (FMDM)



[2] Adapted from Jerden et al. (2015)



- 1-dimensional reactive transport model of a waste package
- Needed for each breached package in the repository at each time point
- Computationally intensive to calculate UO_2 degradation rates
- Surrogate models map inputs to outputs with less computational cost

Training Data

Sampling Input Parameters

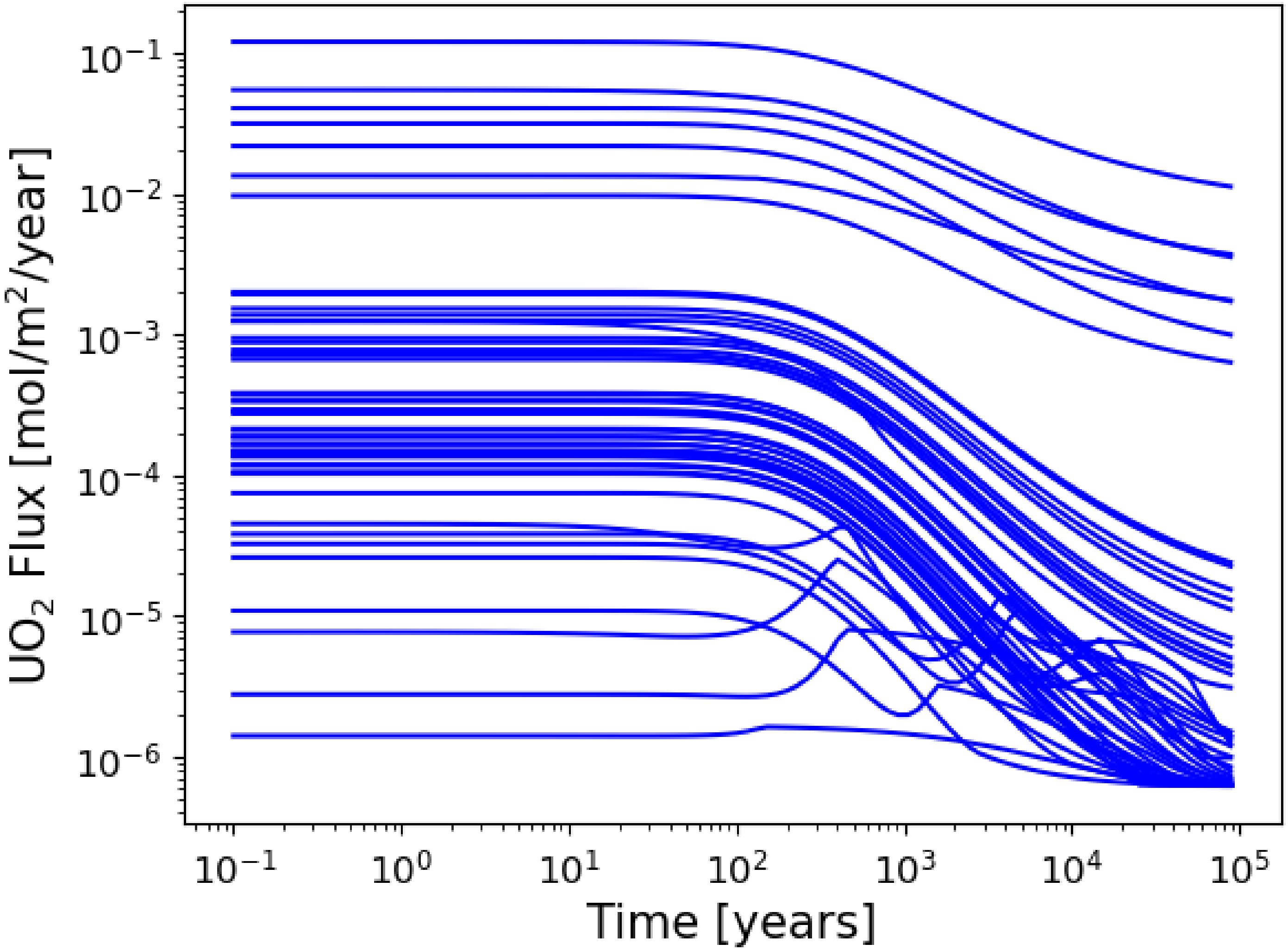
Parameter	Distribution	Min.	Max.
Init. Temp. (K)	Uniform	300	600
Burnup (Gwd/MTU)	Uniform	40	80
Delay Time (years)	Log-uniform	10^2	10^4
Env. CO_3^{2-} (mol/m^3)	Log-uniform	10^{-3}	2×10^{-2}
Env. O_2 (mol/m^3)	Log-uniform	10^{-7}	10^{-5}
Env. Fe^{2+} (mol/m^3)	Log-uniform	10^{-3}	10^{-2}
Env. H_2 (mol/m^3)	Log-uniform	10^{-5}	2×10^{-2}

- Process model input parameters sampled from expected ranges in reservoir simulations to generate training data time-trajectories
- Ranges that span multiple orders of magnitude sampled with log-uniform distribution
- Parameters are externally-imposed

Surrogate Inputs and Outputs

- Environmental CO_3^{2-} and H_2
- Temperature
- Dose rate (function of time and burnup)

UO_2 Degradation (Surface Flux)

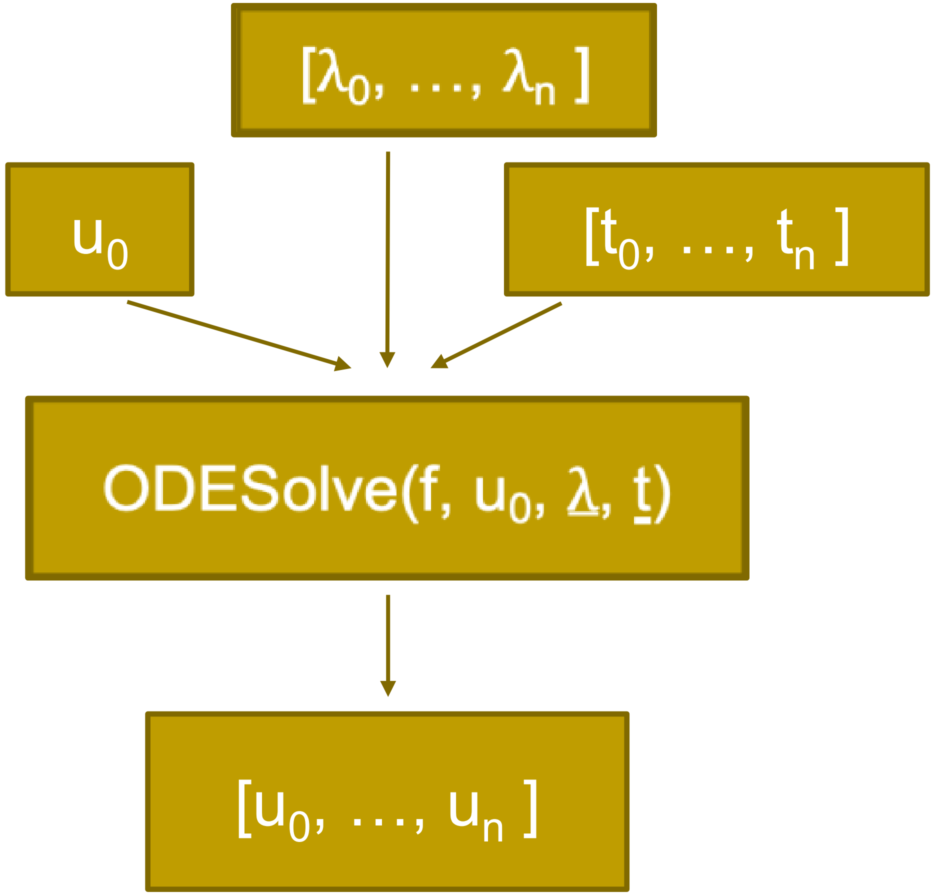


Neural ODEs

Methods

Neural ODEs Background

$$\frac{du}{dt} = f(u, \lambda) = NN$$

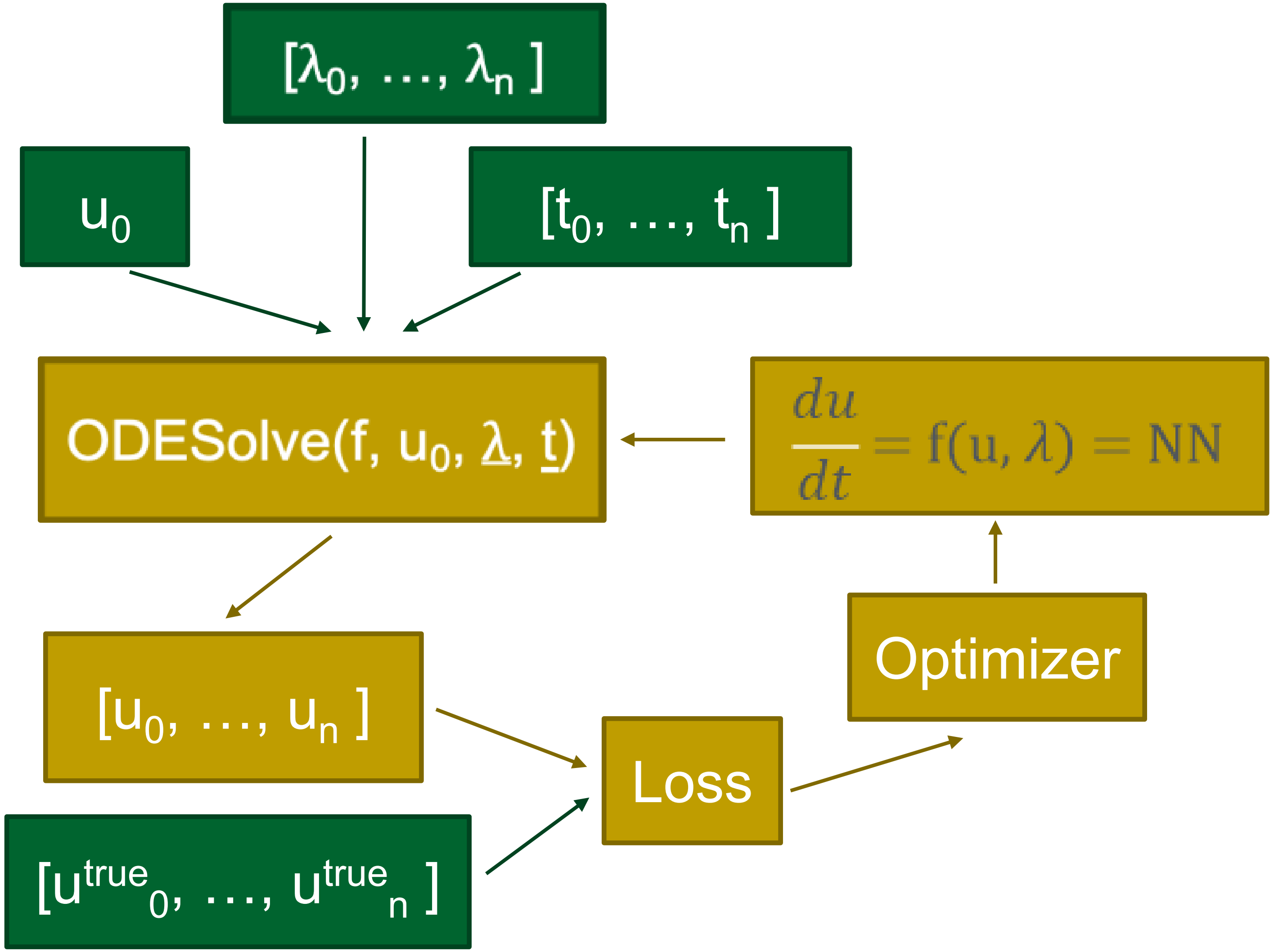


- Neural ODEs approximate the derivative of the system state as a Neural Network
- Predict with ODE Solver

[3] “Neural Ordinary Differential Equations”, Chen et al.

Training Process and Hyperparameters

- Select random points from a trajectory to serve as batch initial conditions
- Hyperparameters to tune:
  - Learning rate
  - Number of layers
  - Number of neurons per layer
  - Number of batches (batch_size)
  - Number of time steps to predict/integrate during training (batch_time)
  - Amount of training data
  - Number of time points to use from training data
  - Choice of numerical ODE solver

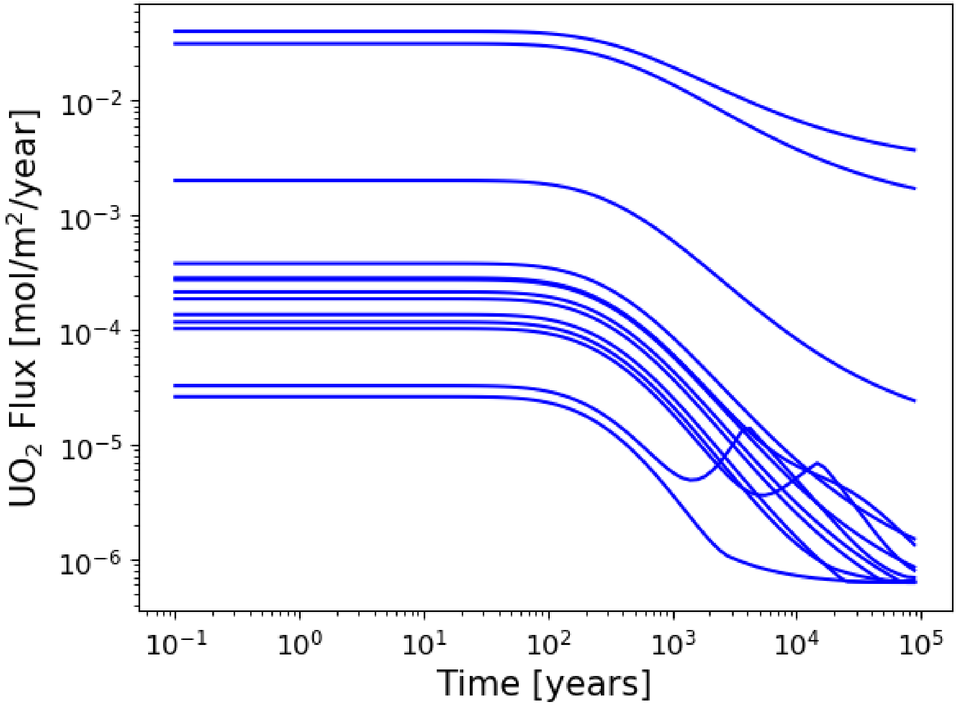


Neural ODEs

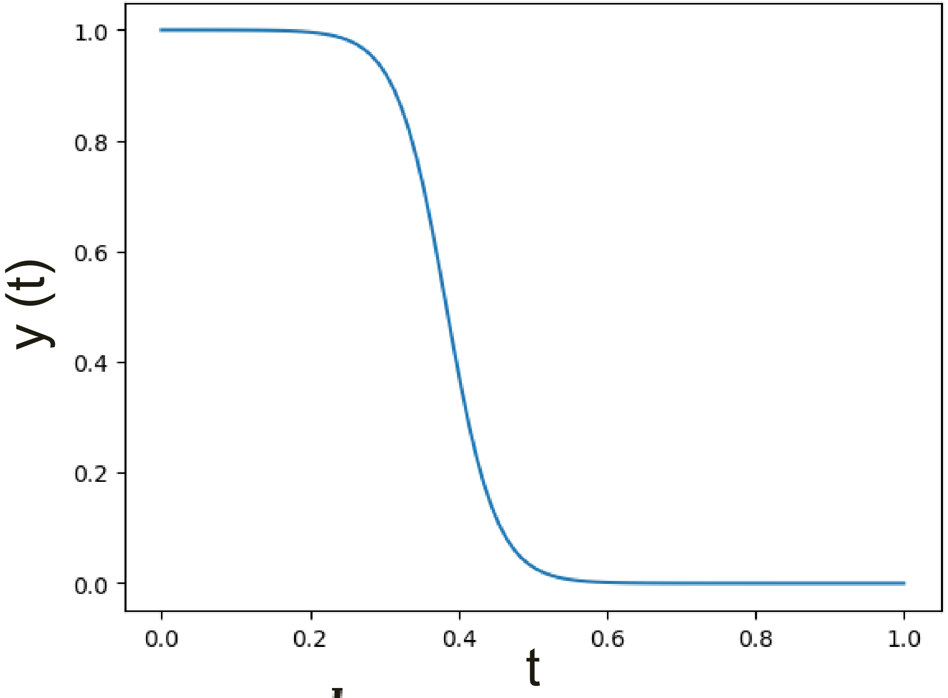
Logistic Decay Toy Problem

Simplify Problem-Solving with Logistic Decay ODE

- Logistic Decay ODE with precise initial conditions mimics plateau-and-decay shape of UO_2 flux trajectories but has known dynamics and no parameter dependence



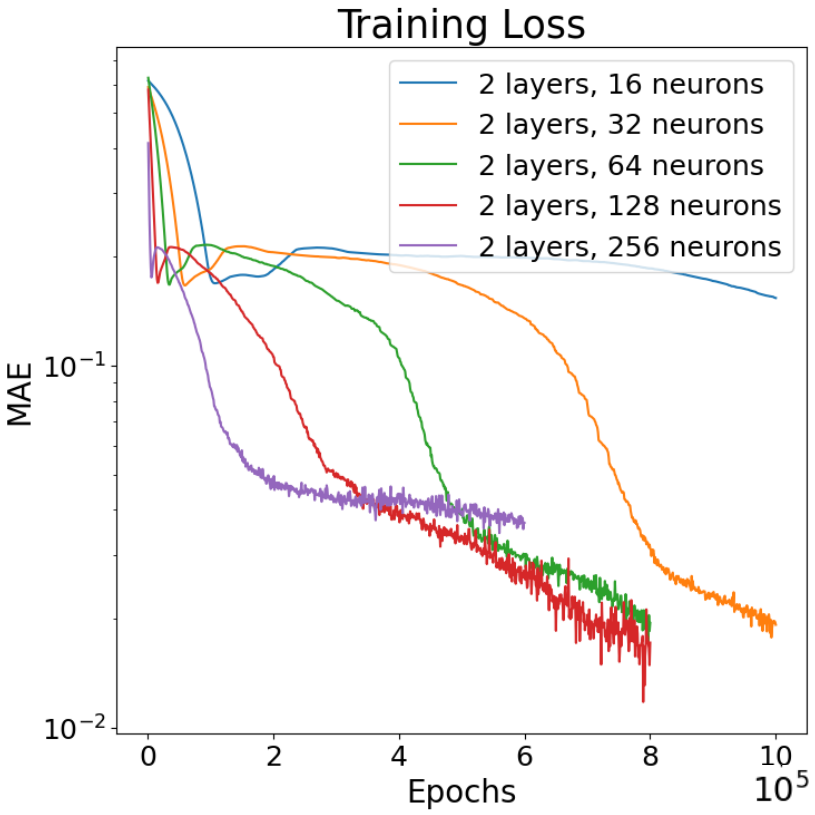
UO_2 Flux



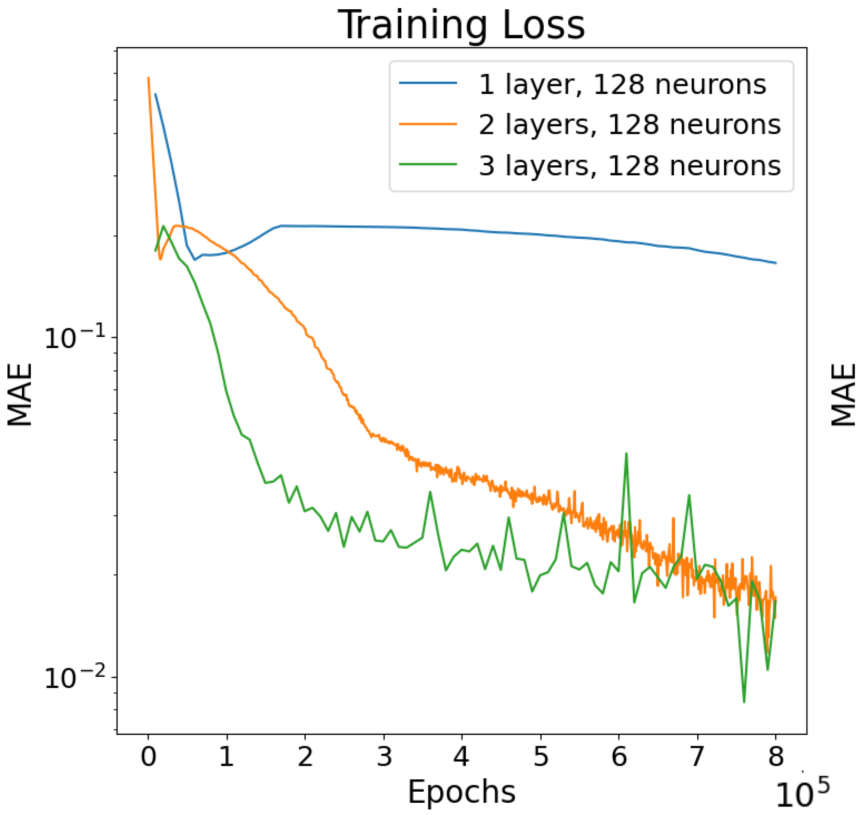
$$\frac{dy}{dt} = -30y(1-y),$$
$$y(0) = 0.99999$$

Experiments with Logistic Decay

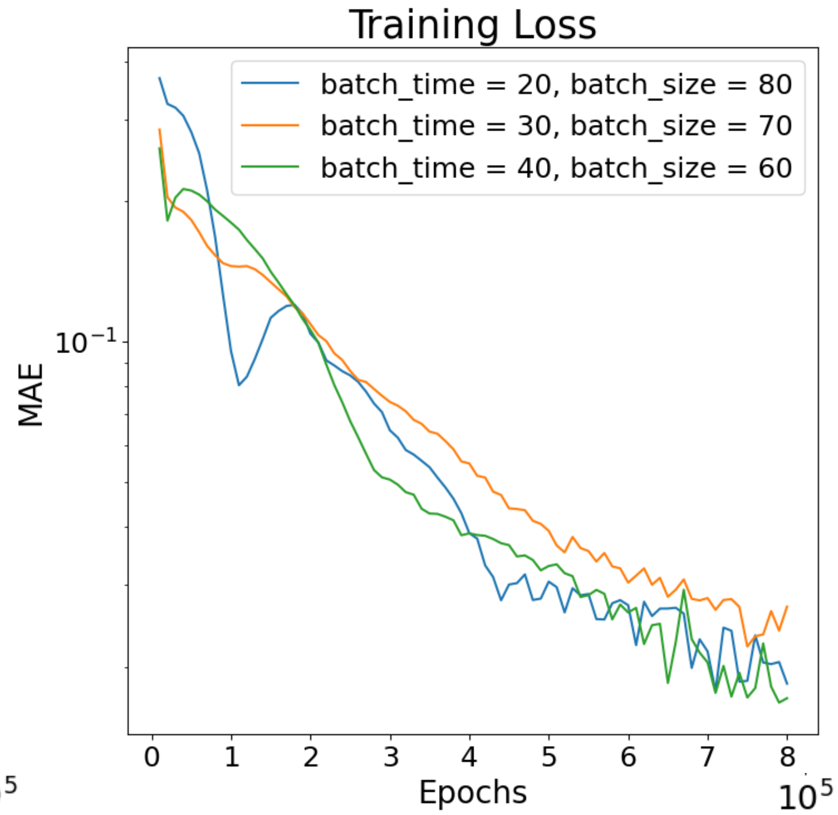
- Dopri5 has best balance of speed and accuracy in torchdiffeq.odeint solvers (with NRMSE = 9.4490e-05 when solving the true RHS of the ODE)
- Precision of the problem requires a learning rate of 1e-6
- Lowest training loss with 2 layers, 128 neurons, batch_time 20, and batch_size 80



batch_time = 40, batch_size = 60



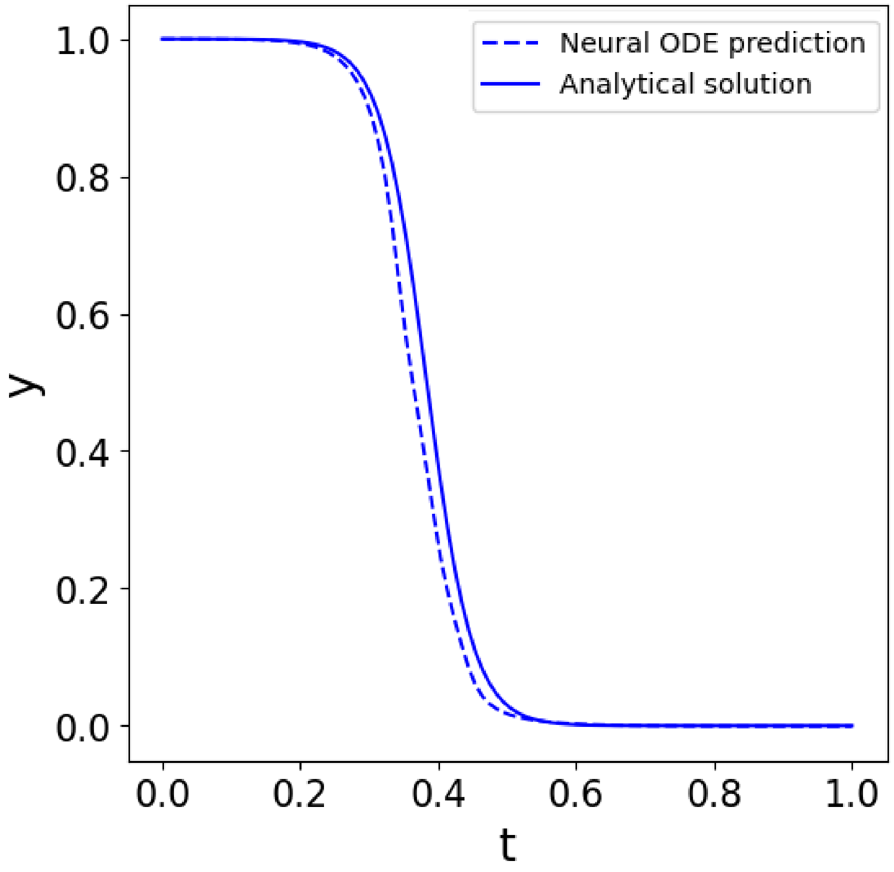
batch_time = 40, batch_size = 60



2 layers, 128 neurons/layer

Logistic Decay Results

- Neural ODEs have difficulty learning a trajectory with an initial plateau
- Very sensitive to initial conditions
- For the best set of hyperparameters, training NRMSE = 0.08323 after 1.2M epochs



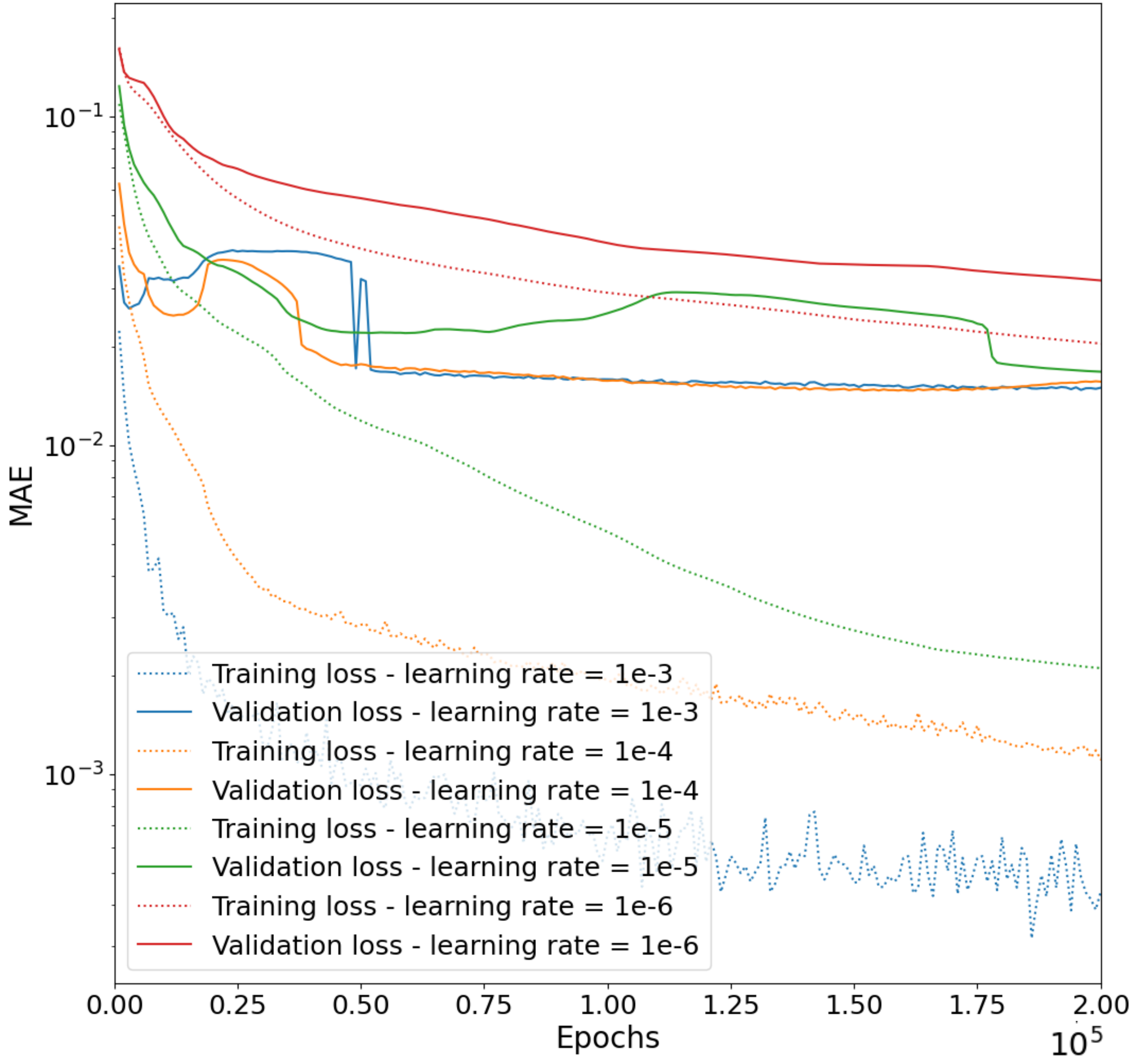
Neural ODEs

FMD Application

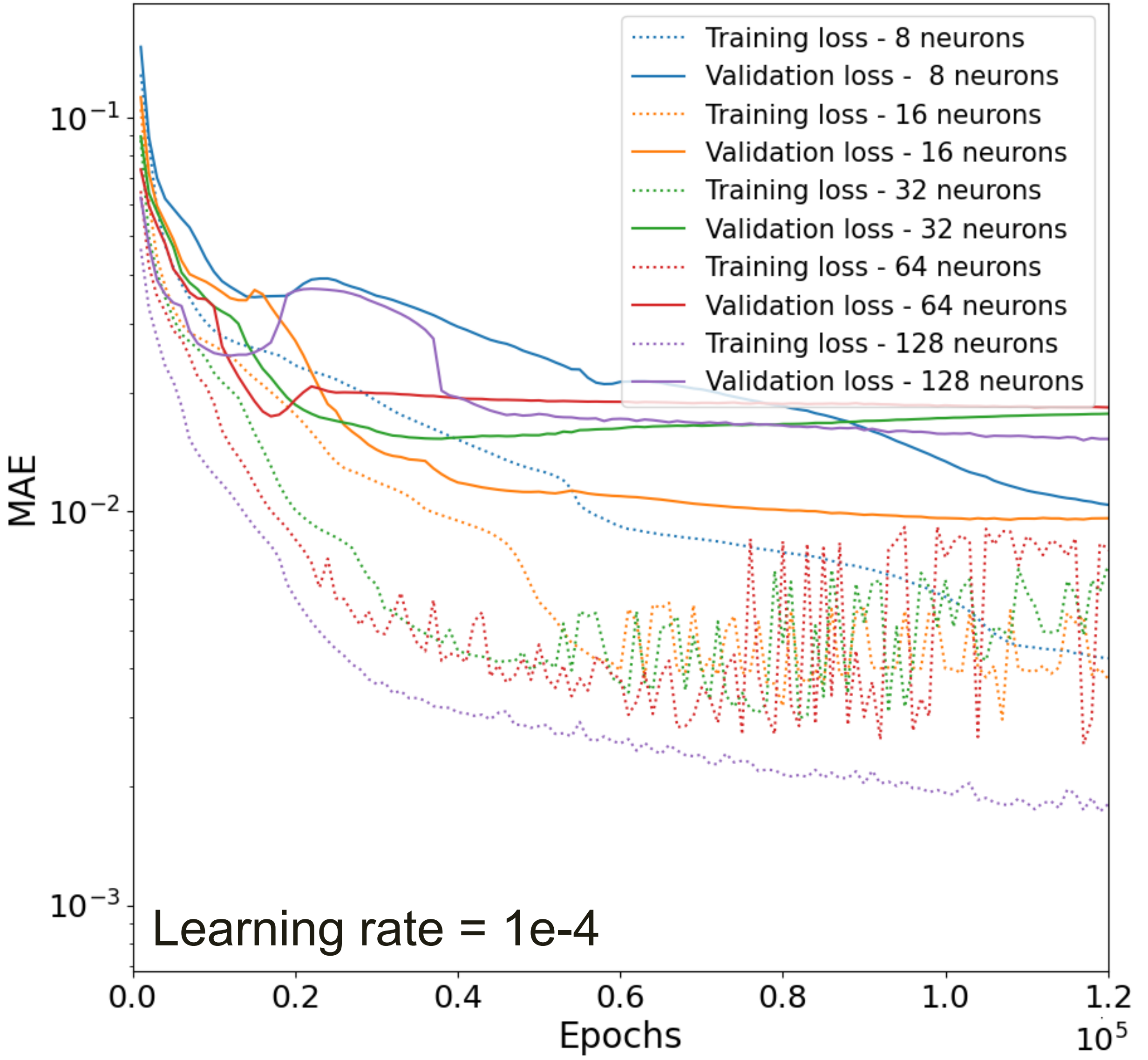
Hyperparameter Tuning: Learning Rate

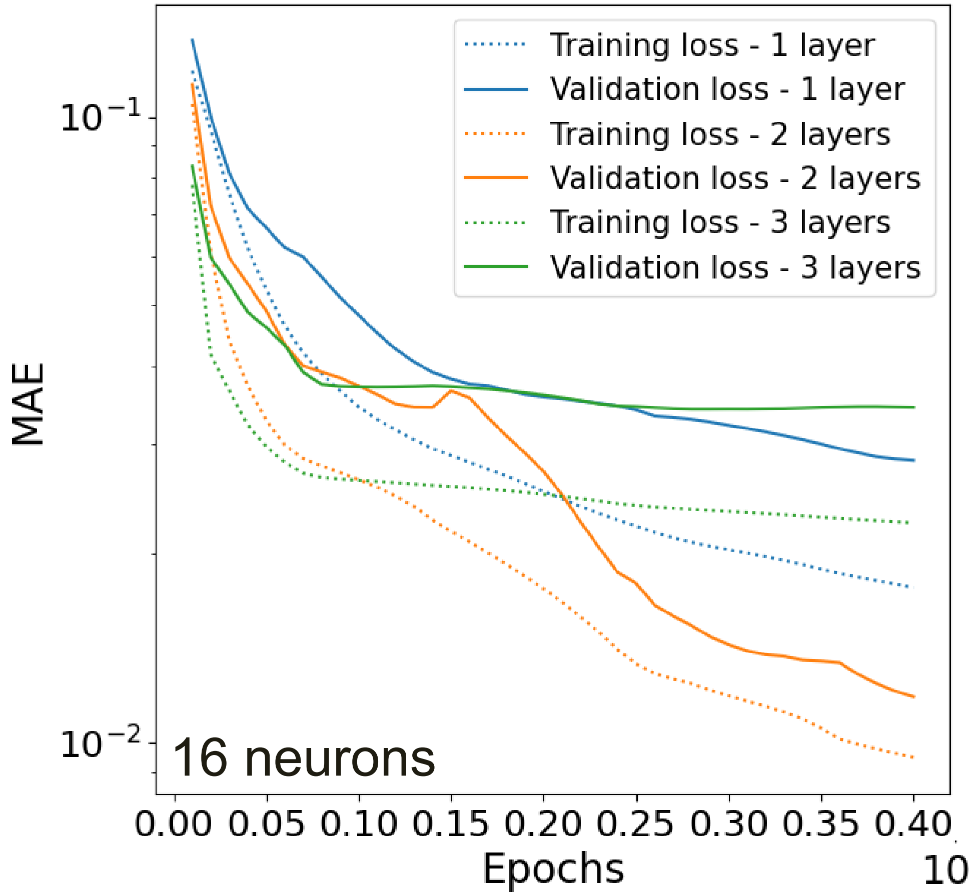
- For preliminary tests, we use
  - torchdiffeq.dopri5 solver
  - 100 trajectories (80 training/ 20 validation)
  - 100 time steps
  - Batch_time = 20
  - Batch_size = 80
  - 2 layers, 128 neurons

- A learning rate of 1e-4 is a good balance of speed, accuracy, and stability



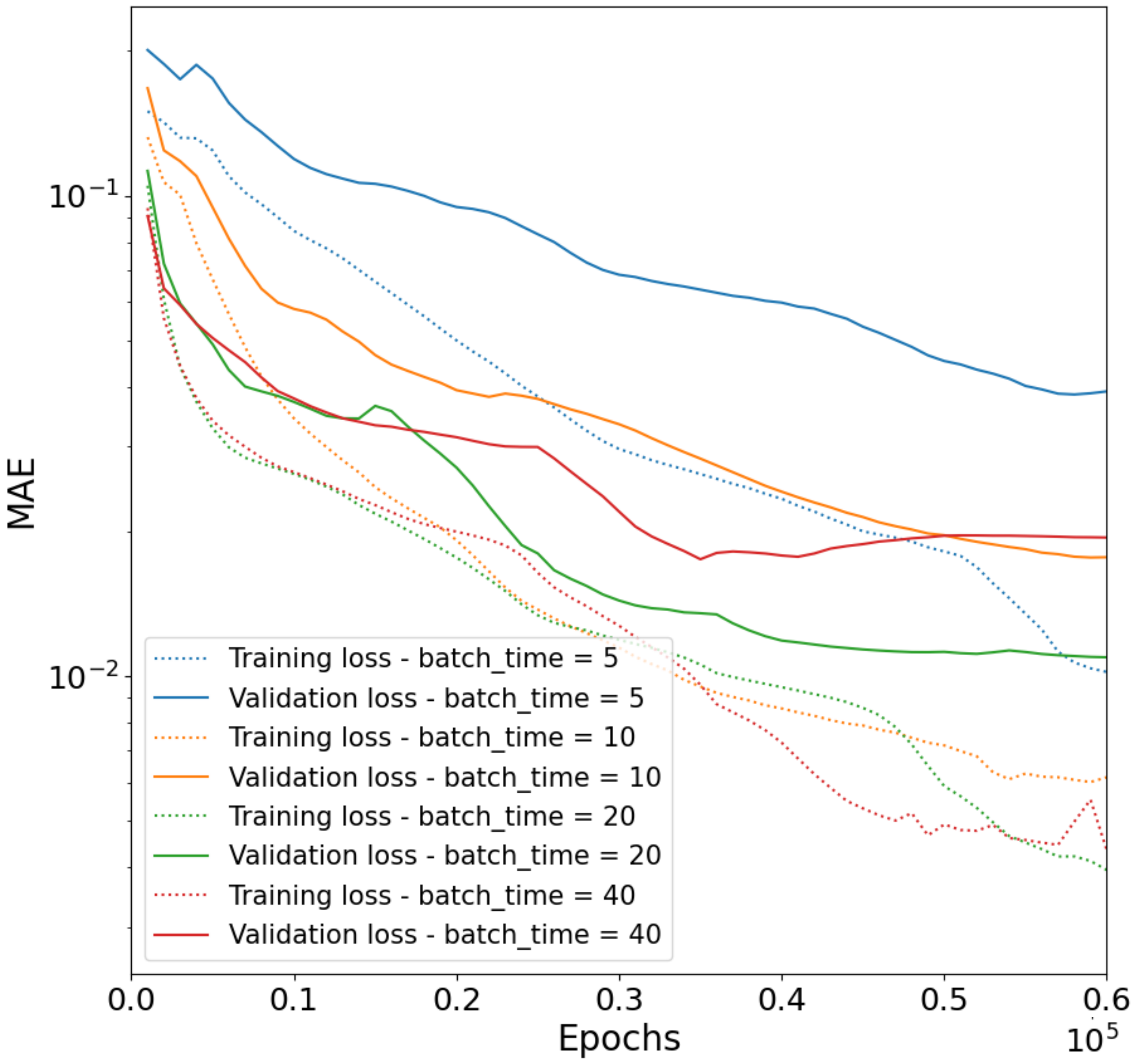
Hyperparameter Tuning: Neurons and Layers





- An architecture of 16 neurons and 2 layers has the lowest validation loss

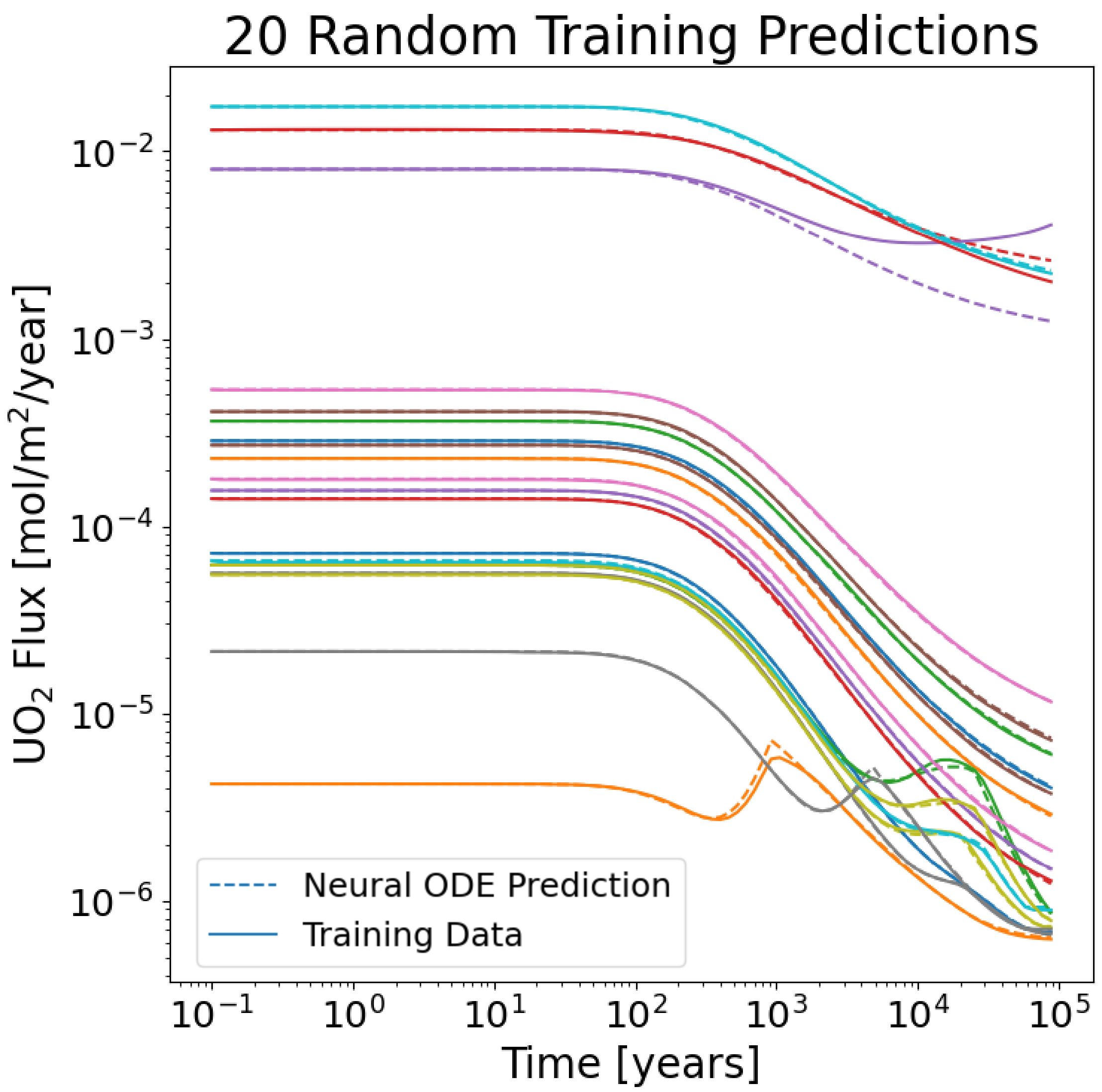
Hyperparameter Tuning: batch_time

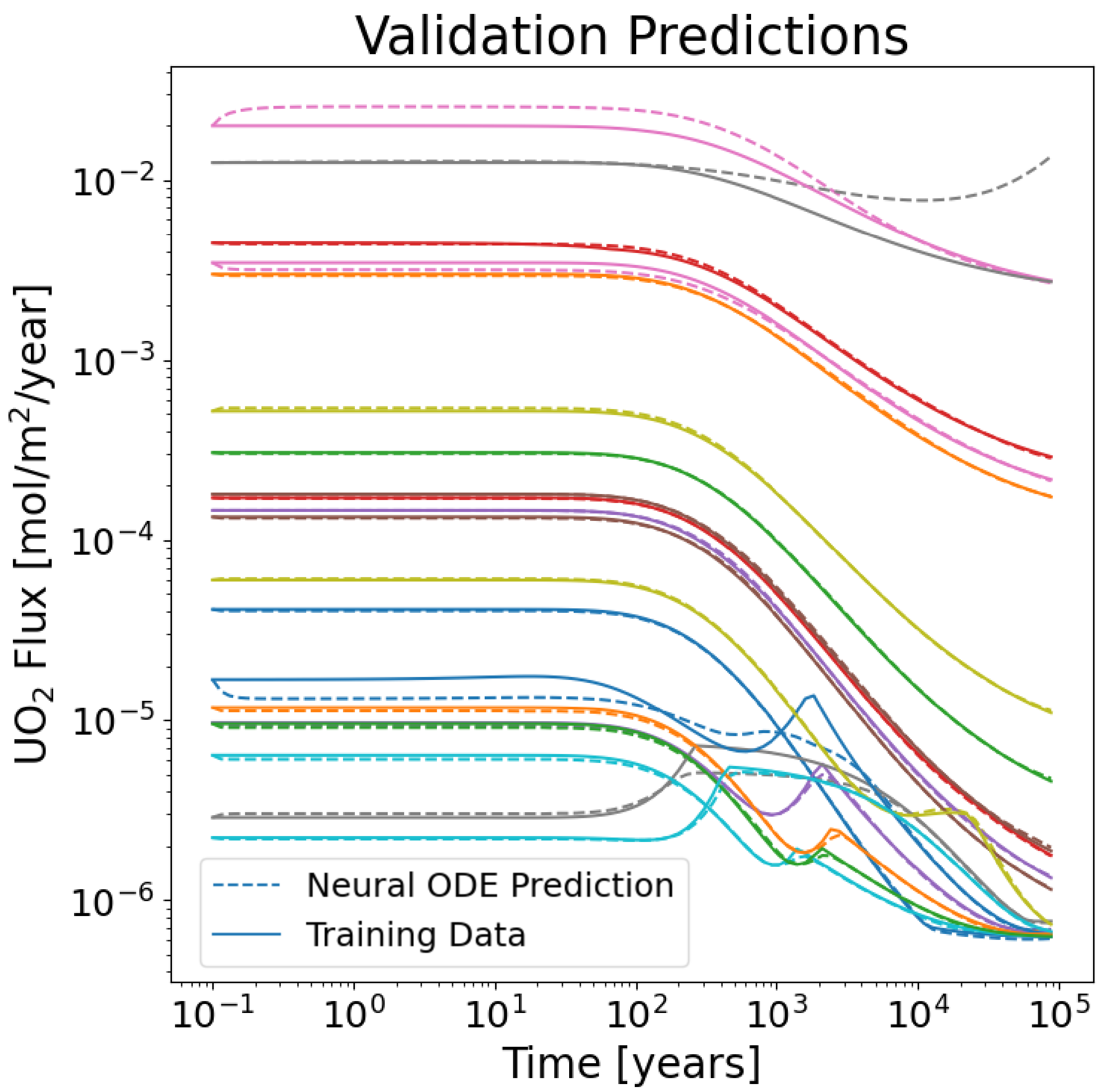


- Batch_time = 20 yields the lowest validation loss

Note: For each batch_time, batch_size = # time steps –

Prediction of FMD Validation Data from Initial Flux





MAE = 0.0072875 NRMSE = 0.066859

MAE = 0.0095912 NRMSE = 0.230

Conclusions and Future Work

Conclusions and Ongoing Work

- Choosing an accurate ODE solver and a large enough batch_time are crucial for learning the timing of the slope transition.
- Training requires over 100,000 epochs and may involve local minima in the loss.
- Neural ODEs have shown the ability to learn the dynamics of the FMDM data and predict a time trajectory from its initial condition, and therefore, have potential as a surrogate.
- Ongoing Work
  - Further Hyperparameter tuning of
    - Number of trajectories of training data
    - Number of time steps in training data
    - Choice of numerical solver
  - Cross-validation for hyperparameter tuning
  - Evaluation on testing data

References

1. Freeze, G., E.J. Bonano, P. Swift, E. Kalinina, E. Hardin, L. Price, S. Durbin, R. Rechard, and K. Gupta, “Integration of the Back End of the Nuclear Fuel Cycle.” SAND2021-10444. Sandia National Laboratories, Albuquerque, New Mexico (2021)

2. J. Jerden, K. Frey, and W. Ebert, “A Multiphase Interfacial Model for the Dissolution of Spent Nuclear Fuel,” Journal of Nuclear Materials, 462, 135, https://doi.org/10.1016/j.jnucmat.2015.03.036 (2015)

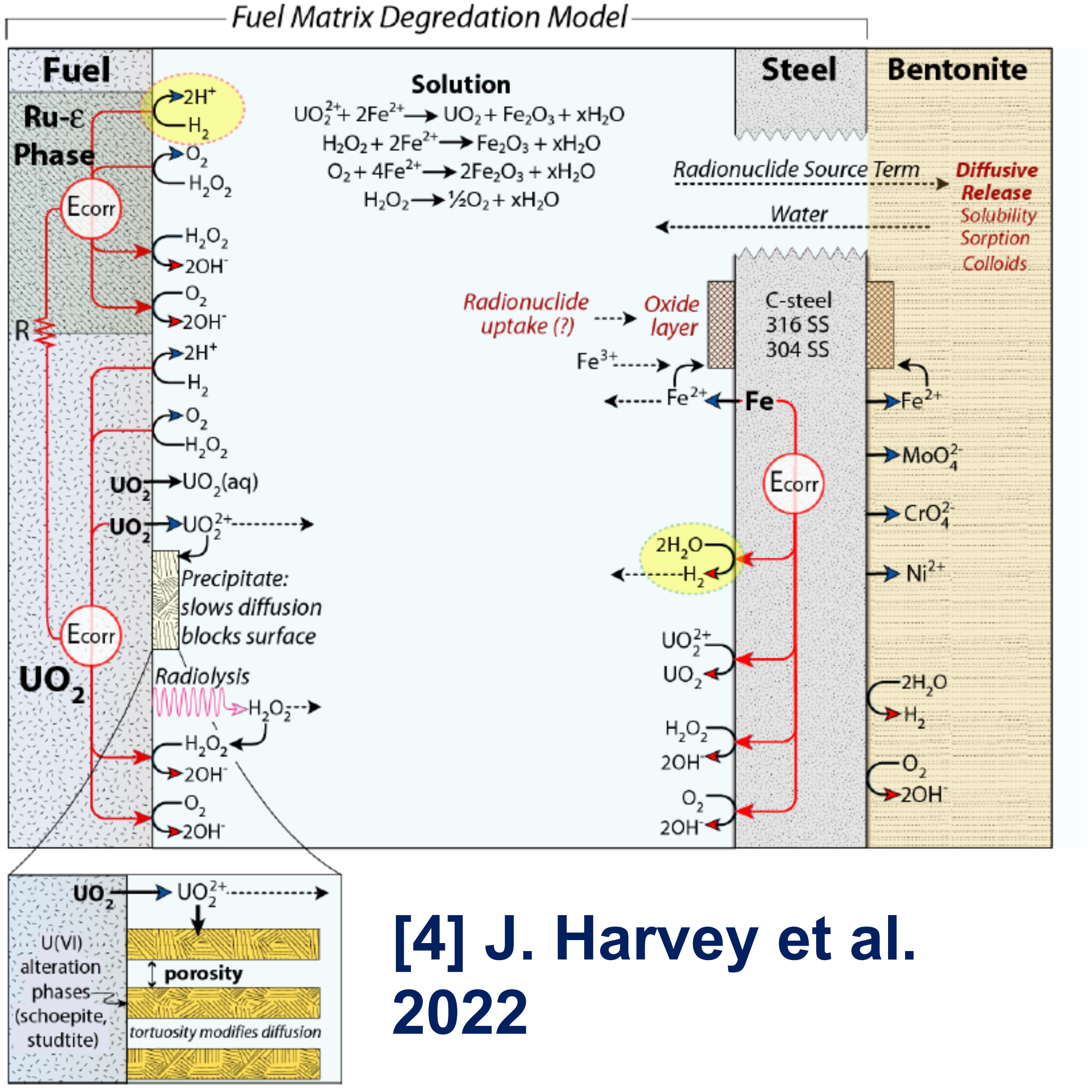
3. Chen, Ricky T. Q., Yulia Rubanova, Jesse Bettencourt, and David K. Duvenaud, “Neural Oridinary Differential Equations.” Advances in Neural Information Processing Systems 31 (2018).

4. J. Harvey et al.: Development of an Efficient Version of the Fuel Matrix Degradation Model, IHLRWM 22 (2022)

Additional Materials

The Fuel Matrix Degradation (FMD) process model computes the degradation rate of spent nuclear fuel

- 1D reactive-transport model (diffusion only)
- Chemical (slow) and oxidative (fast) dissolution of UO_2 matrix
- Hydrogen peroxide production via alpha-radiolysis
- Precipitation and dissolution of U(VI) (i.e., schoepite) corrosion layer at the fuel surface
- Arrhenius temperature dependence
- Complexation of uranium with carbonates
- Hydrogen as an oxidation sink (focused on fuel interface)
- Logarithmic spatial discretization for enhanced accuracy near the solid interfaces



[4] J. Harvey et al. 2022

Surrogate FMD models can alleviate cost of UO_2 flux computation in probabilistic repository assessments

Input Parameters

Parameter database

Computational Support

Uncertainty Sampling and Sensitivity Analysis

DAKOTA

Explore and predict with confidence.

Processing

VoroCrust

python

dfnWorks

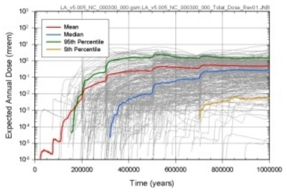
Visualization

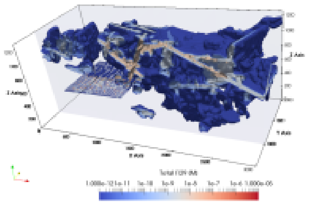
ParaView

Parallel Visualization Application

VisIt

Results





Fuel Matrix Degradation Model (FMDM)

Offline, MATLAB

FMDM Surrogates:

- ✓ PFLOTRAN Coupled
- ✓ HPC Speed and Scale
- ✓ Fortran

Multi-Physics Simulation and Integration

PFLOTRAN

Models

Source Term and Engineered Barrier System Evolution Model

Inventory
Decay, ingrowth
Waste form degradation
Waste package degradation
Radionuclide release
Thermal, mechanical
Gas generation

Biosphere Model

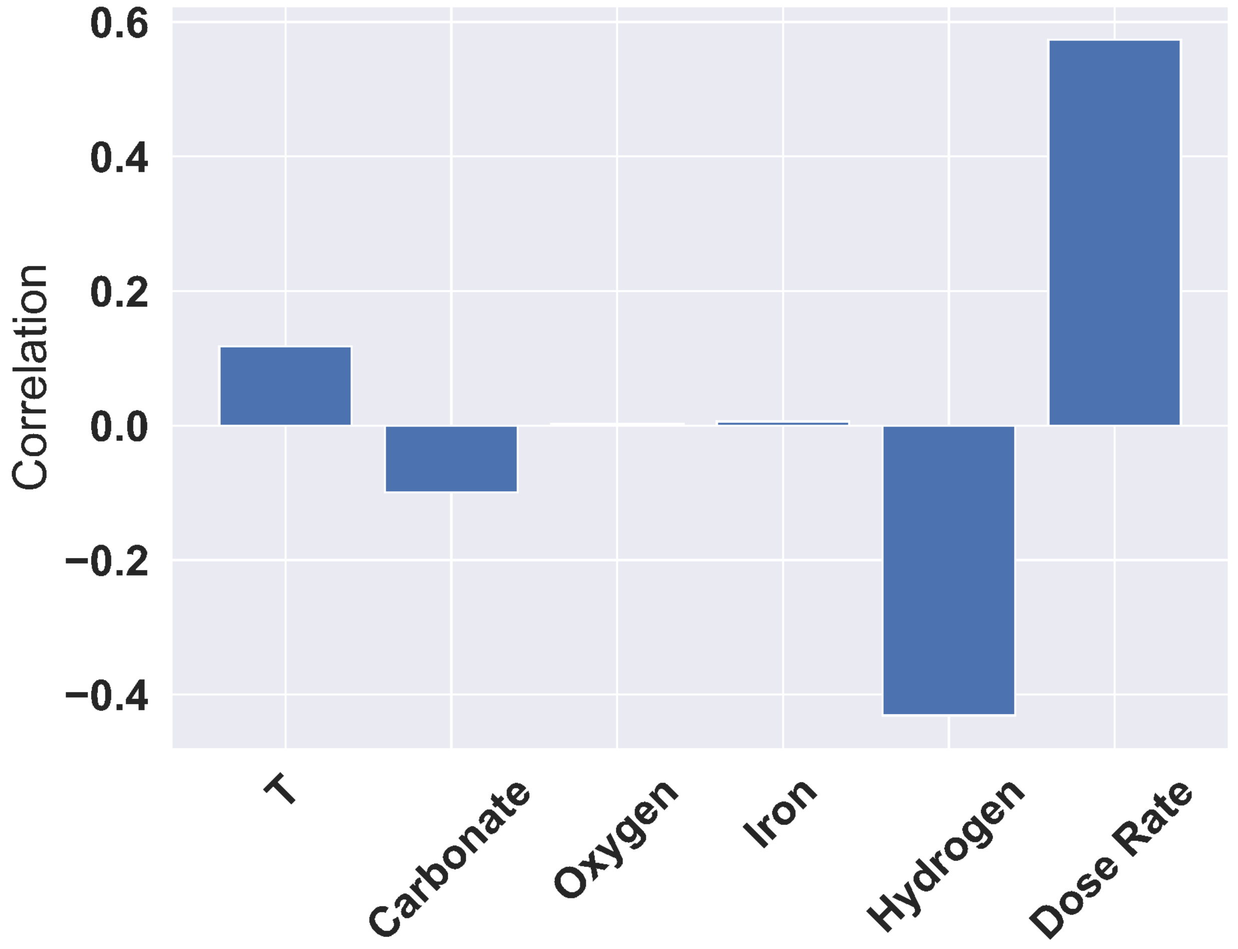
Exposure pathways
Uptake/ transfer
Dose calculations

Flow and Transport Model

Advection, diffusion, dispersion
Discrete fracture networks
Multiphase flow
Sorption, solubility, colloids
Isotope partitioning
Decay, ingrowth
Thermal effects
Chemical reactions

Model inputs that do not impact the fuel degradation rate much can be dropped

- Correlation between fuel degradation rate and O_2, Fe^{2+} is very small



Data conditioning improves the quality of the training data

- Remove FMD process model runs that are physically unrealistic
  - Runs that do not finish
  - Runs that stagnate at late time
  - Runs with Corrosion Layer Thicknesses that exceed physical domain size
  - Runs with UO_2 (aq) over 2 times the solubility concentration

- Log-transform data

- Scale between -1 and 1

A variety of metrics evaluate different elements of the surrogate model accuracy

- (Normalized Root) Mean Squared Error
  - Good metric for engineering purposes

$$mse = \frac{1}{N}\sum_{i=1}^{N}\left(y_{pred,\,i} - y_{true,\,i}\right)^2$$

$$nrmse = \frac{\sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(y_{pred,\,i} - y_{true,\,i}\right)^2}}{\frac{1}{N}\sum_{i=1}^{N} y_{true,\,i}}$$

- Mean Absolute Percentage Error
  - Highlights errors in small values

$$mape = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{y_{pred,\,i} - y_{true,\,i}}{y_{true,\,i}}\right| \times 100$$

- Mean Absolute Error
  - Not as sensitive to outliers

$$mae = \frac{1}{N}\sum_{i=1}^{N}\left|y_{pred,\,i} - y_{true,\,i}\right|$$