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REACTIVE TRANSPORT MODELS AND METAMODELS FOR THE GEOCHEMICAL EVOLUTION IN A HLW REPOSITORY

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OUTLINE

- **Introduction**
- **High-fidelity reactive transport models for lab corrosion tests**
- **CORE code improvements applied to HLW repository**
- **Geochemical machine learning benchmark:**
  - Metamodel for a corrosion reactive transport model
  - Metamodel for a uranium system
- **Conclusions & Future work**

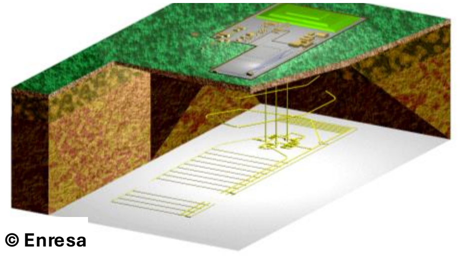
INTRODUCTION

- **This work is developed by the UDC within EURAD2 project-WP HERMES:**
  - **High-Fidelity models**: Enhance Thermal-Hydraulic-Chemical (THC) models for key iron/bentonite interfaces.
  - **Machine Learning Surrogates:** Develop fast ML metamodels to replace computationally heavy geochemical solvers.
  - **Repository scale simulation:** Model long-term geochemical evolution in granite disposal cells over thousands of years.

Laboratory FeMo test

HLW repository

Deep geological repository



Multibarrier concept:

- Waste
- Fe canister
- Bentonite buffer
- Granite

Chemical barrier

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Engineered barrier

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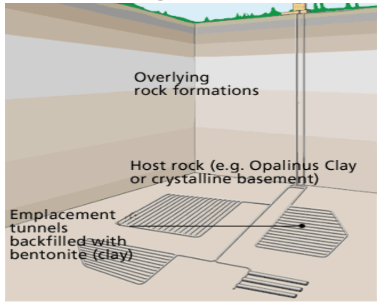
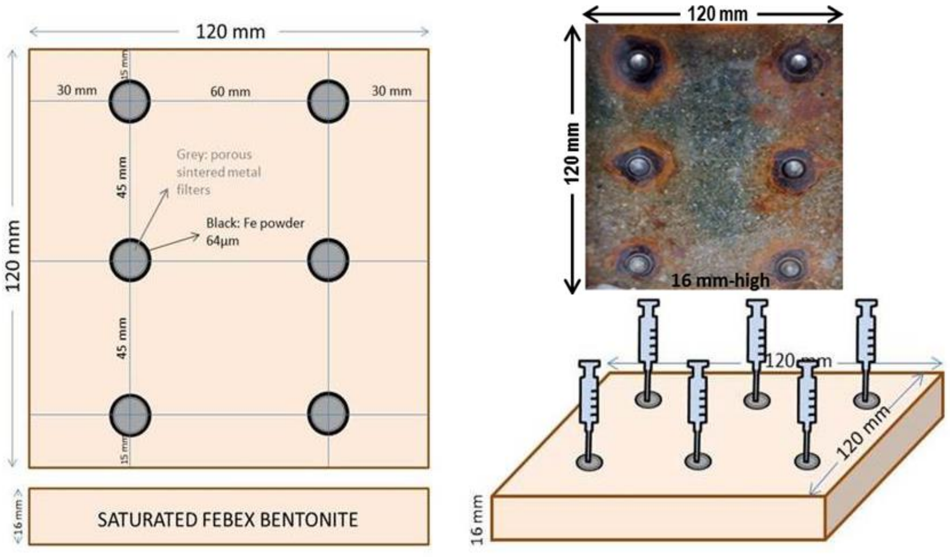
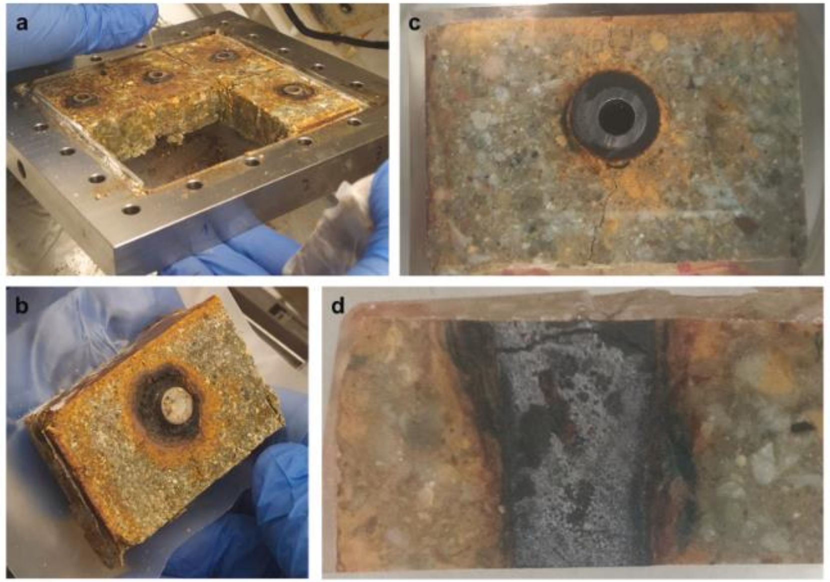
Geological barrier



FEMO CORROSION TEST

- **Laboratory FeMo test description (Turrero et al., 2026)**
  - Compacted bentonite block (120 mm × 120 mm × 16 mm) performed by CIEMAT/UAM
  - Stainless-steel sinter surrounded by iron powder
  - Fe powder particle size of 64 and 240 µm
  - Compacted FEBEX bentonite
  - Hydrated with granite-type water ($CaHCO_3$) through the sinters
  - Corrosion under saturated and ambient conditions
  - Duration of 15 years
  - Experimental data shows magnetite, goethite and siderite precipitation in the iron powder and iron/bentonite interface (Turrero et al., 2026)

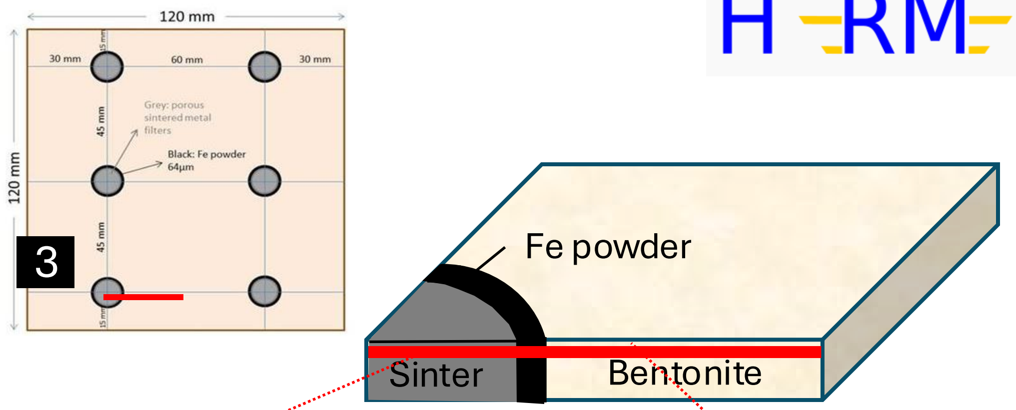


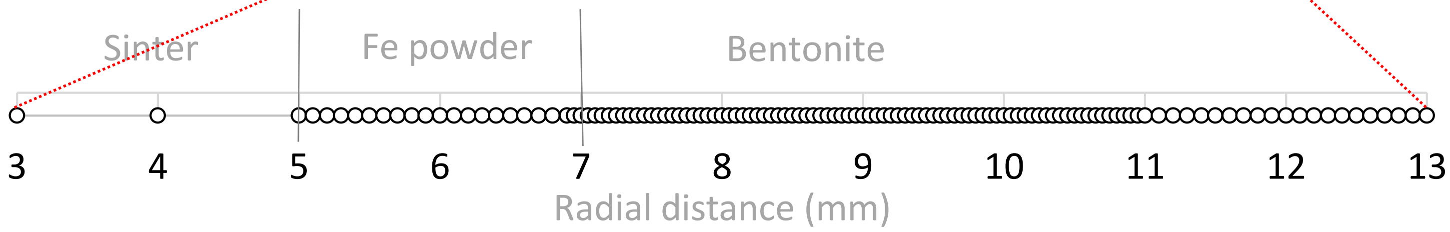


Turrero, M.J., Torres, E., Martín, P.L., Fernández, R., Ruiz, A.I., Ortega, A., Garralón, A., Notario, B., Mota, C., Cuevas, J.F. (2026). Long-term iron-bentonite interaction in a water-saturated room-temperature environment: Advancing for safety of metal waste containment in the geosphere. Surfaces and Interfaces 82, 108466.

FEMO CORROSION TEST

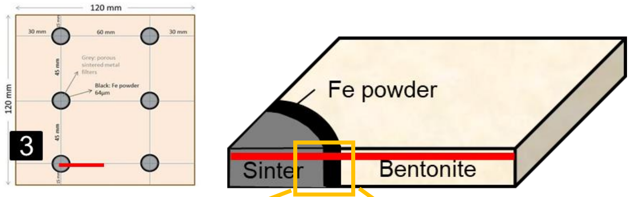
- **Numerical model description**
  - Model of the section 3 (64 µm iron powder grain size)
  - Isothermal and saturated conditions
  - 1D mesh porous media: sinter, Fe powder and bentonite
  - Geochemical reactions:
    - Aqueous complexation
    - Acid-base
    - Redox
    - Cation exchange
    - Surface complexation
    - Mineral dissolution/precipitation
  - Anaerobic corrosion rate.
    - Sinter: very low corrosion (0.02 µm/year)
    - Fe powder: 0.35 µm/year
    - Fe powder/bentonite interface: 0.45 µm/year
  - Calibration of the specific surface areas of the minerals to fit the laboratory observations

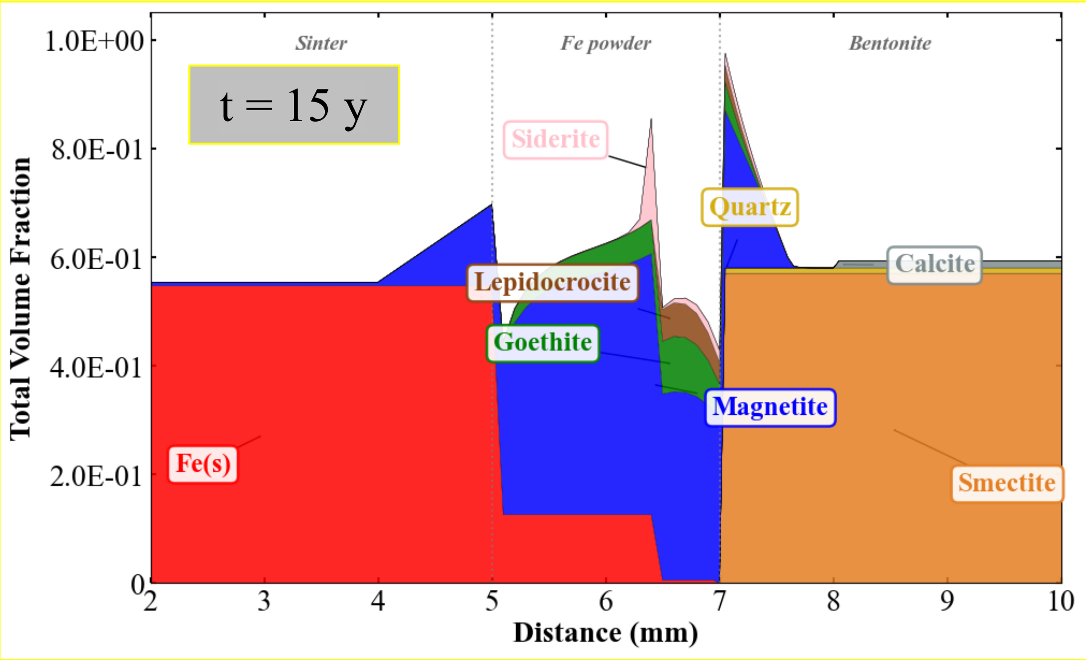


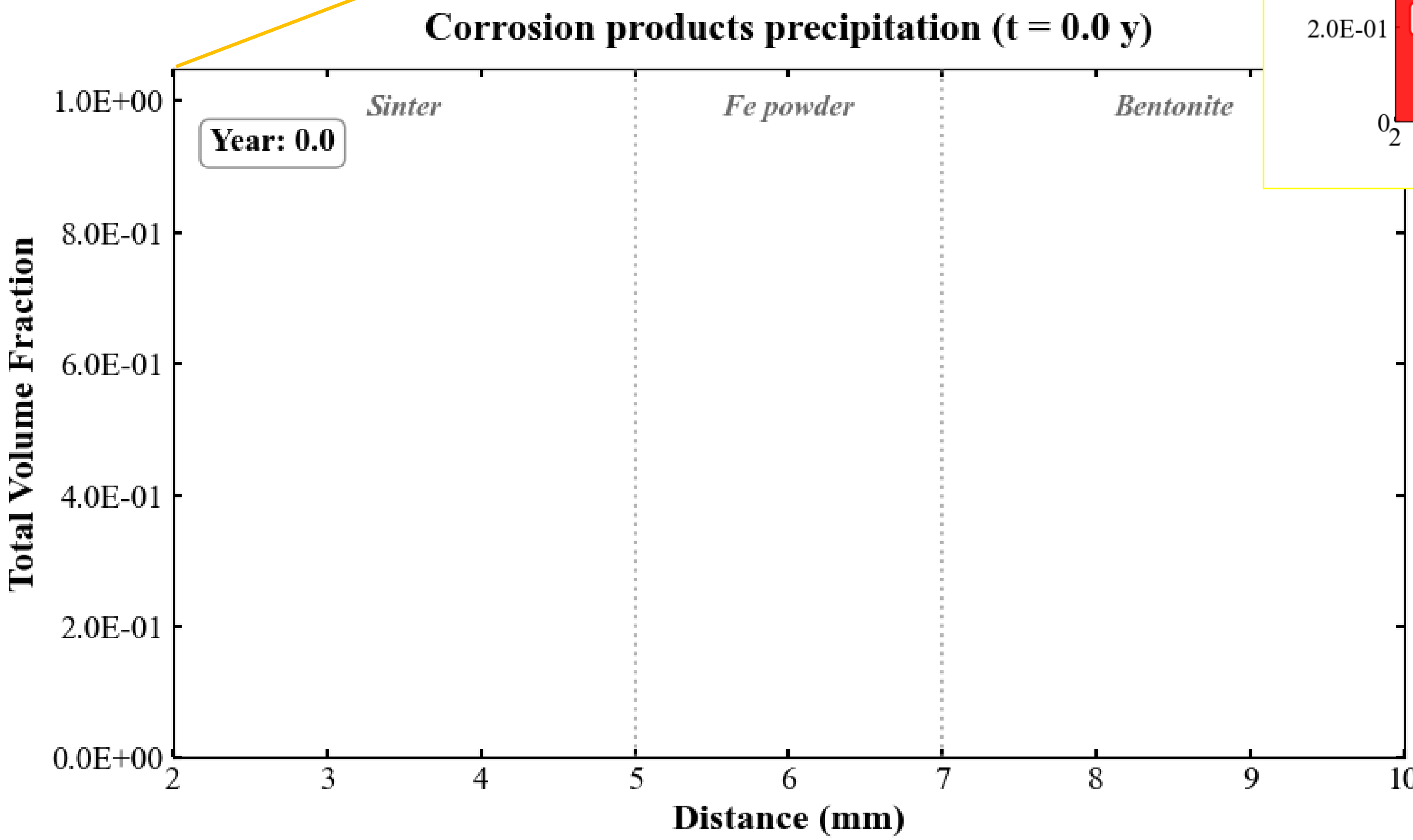


FEMO CORROSION TEST

- Numerical model results: Mineral volume fraction



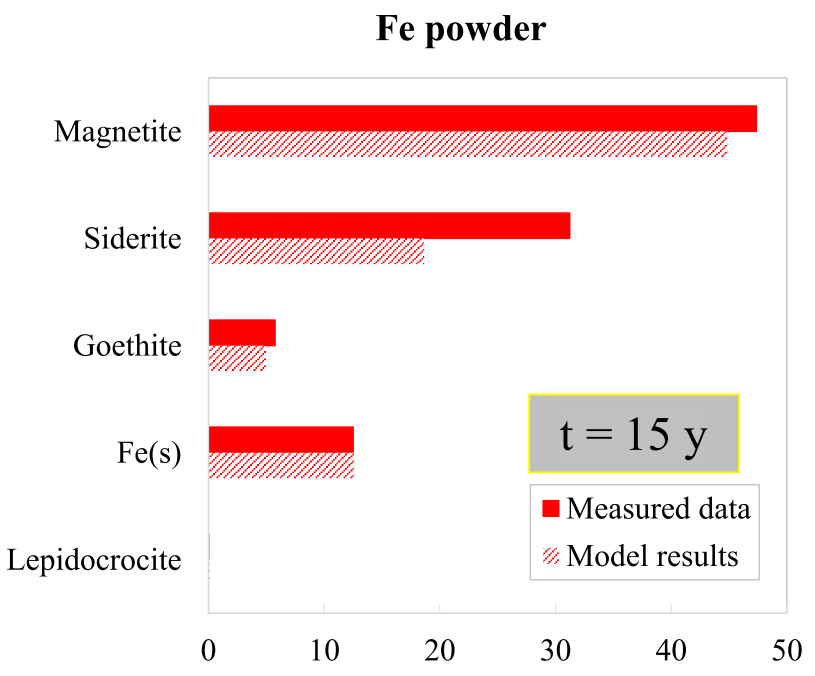


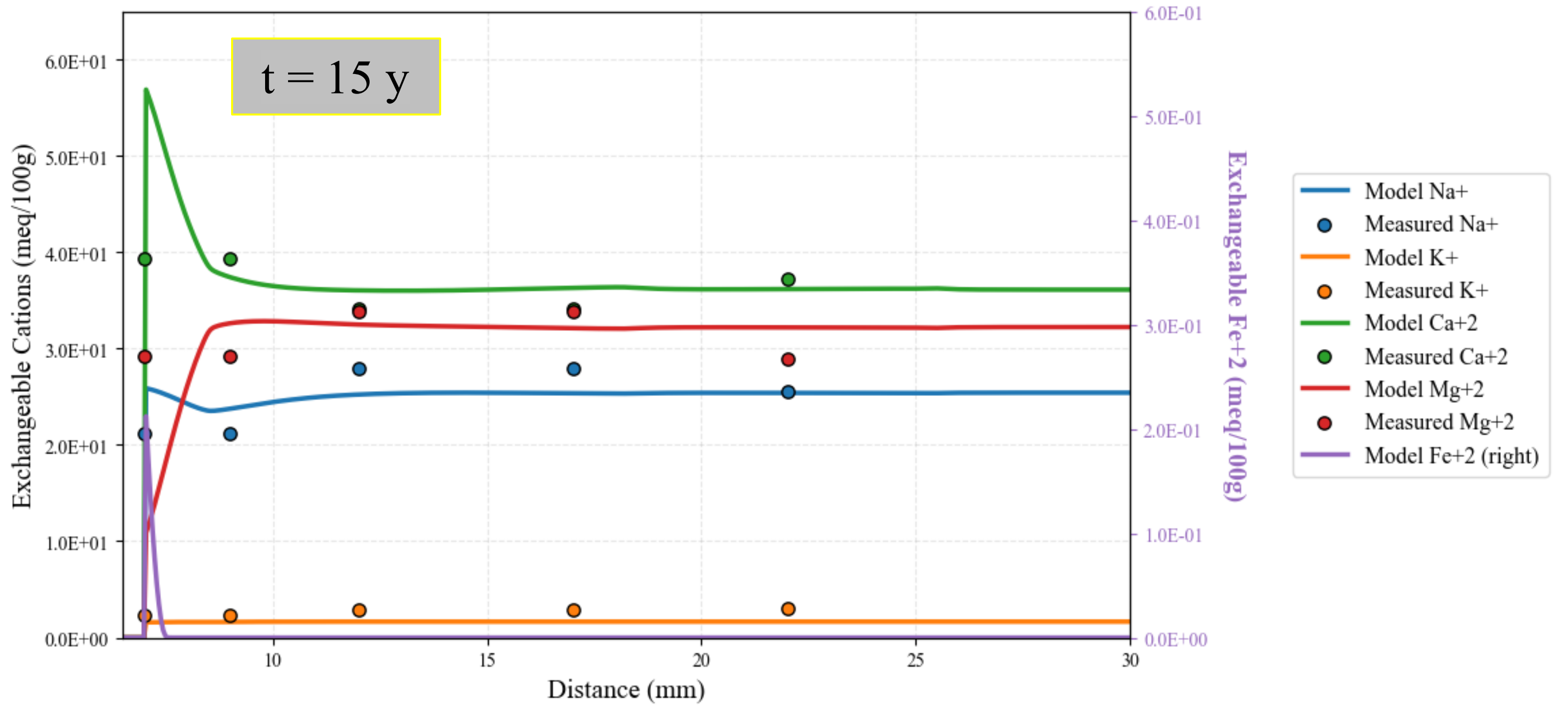


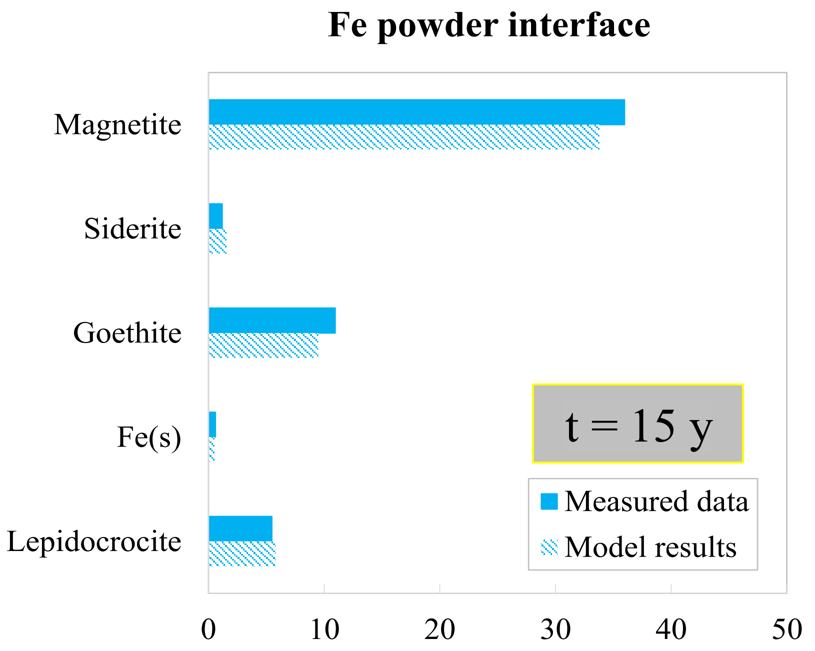
- Fe(s) dissolves in Fe powder and more in the interface
- Magnetite:
  - The main corrosion product
  - Precipitates more in the inner Fe powder
- Goethite and lepidocrocite precipitates more in the interface
- Siderite precipitates in the Fe powder when the HCO_3 arrives

FEMO CORROSION TEST

- **Numerical model results: Comparison with the measured data**
  - Model results fit the volume fraction measured data
  - Model results fit the trend of cation exchange, except at the interface

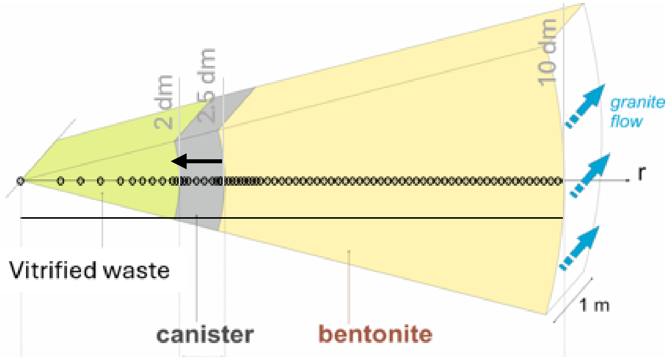


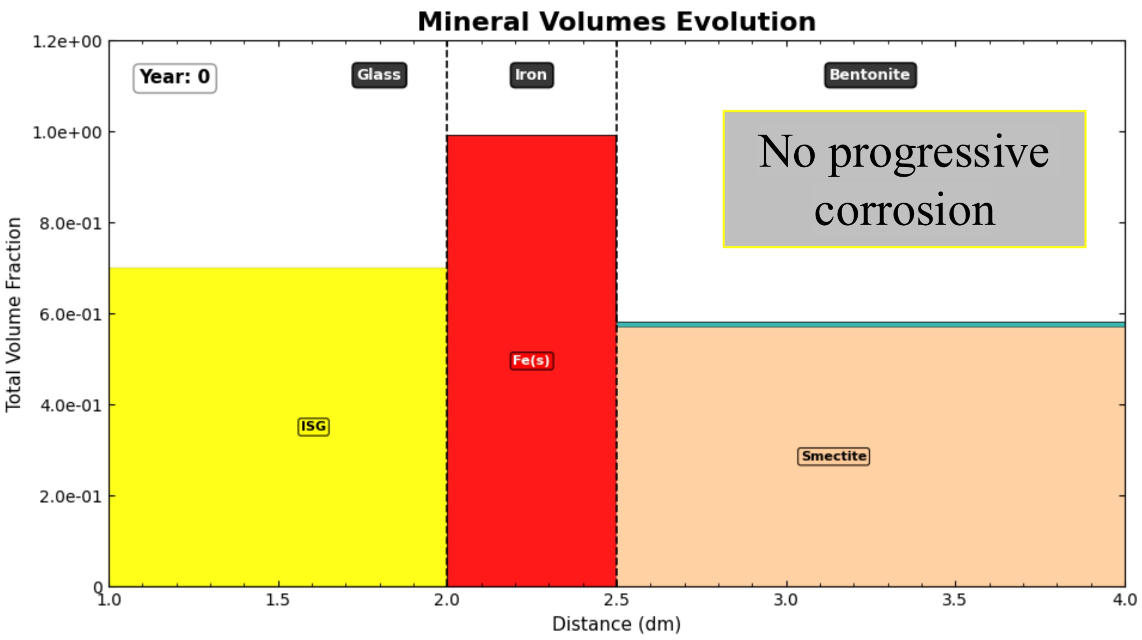


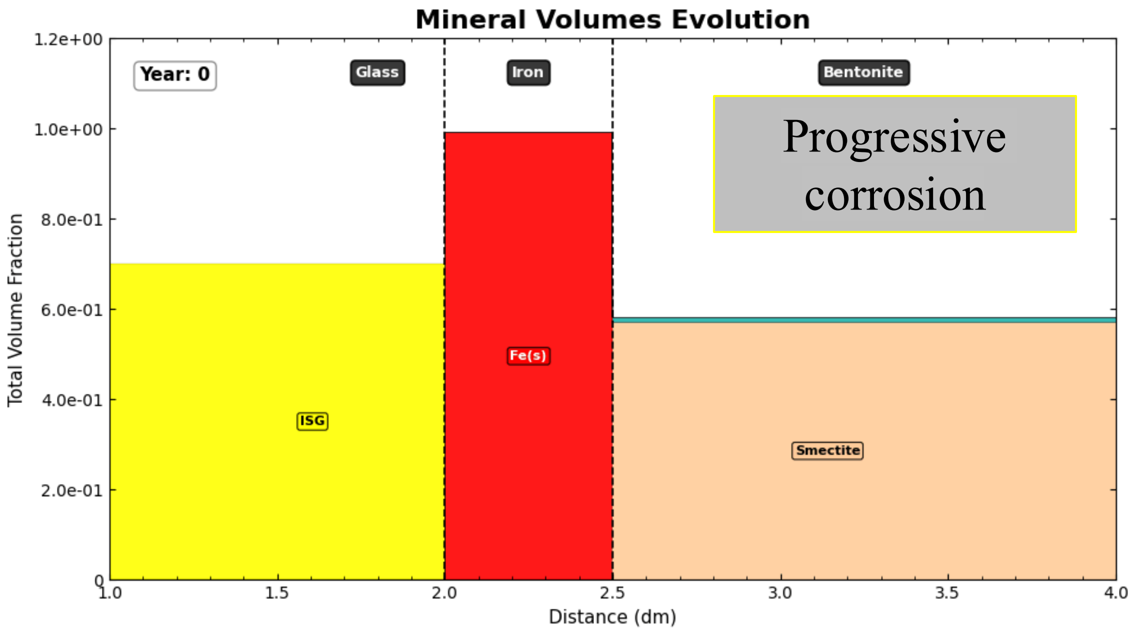


CORE^{2D}V5 CODE IMPROVEMENTS: PROGRESSIVE CORROSION AND PASSIVATION LAYER

- **Progressive corrosion:**
  - Iron corrodes node-by-node rather than all at once, providing realistic spatial front propagation in disposal cells.
- **Magnetite passivation film:**
  - The thickness of the magnetite film is calculated on the assumption that the magnetite precipitates uniformly across the entire surface
  - We calculates uniform magnetite film growth thickness, updating effective diffusion and retarding corrosion rates over time.
- **Application to a HLW repository**
  - Integrated into EURAD-ACED model to simulate long-term geochemical evolution at disposal cell scale.





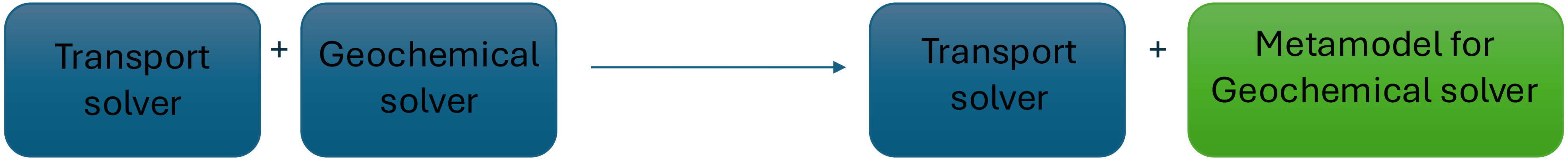


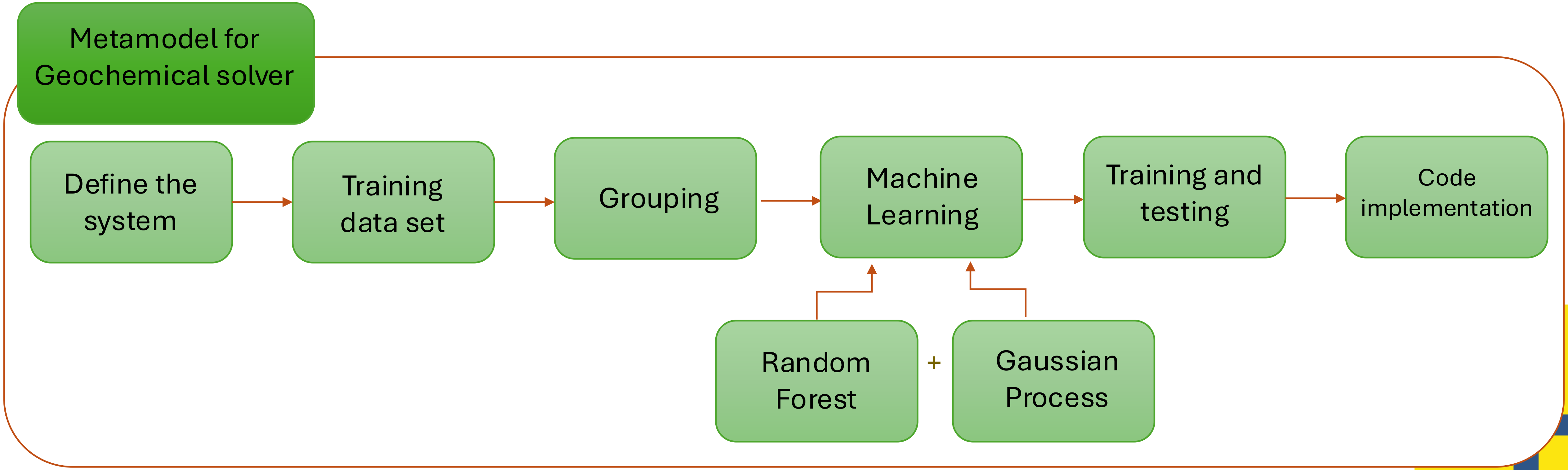
METAMODELS FOR REACTIVE TRANSPORT

- **Motivation:**
  - Reactive transport models require a lot of CPU time mostly for solving the geochemical reactions
    - 24 hours for the FeMo corrosion test
    - More than 10 days for the full long term HLW repository
  - Metamodels and surrogate models provide:
    - Approximate very efficient solutions
    - Emulate the high-fidelity reactive transport simulations
    - Reduce significantly the CPU times
  - Here we present metamodels which could replace the geochemical solver for a chemical system involving canister corrosion and interactions with clay buffer
  - On going work within HERMES WP GEOML benchmark initiative

METAMODELS FOR REACTIVE TRANSPORT

- **Strategy for the metamodel:**





METAMODELS FOR CORROSION

- **Sampling data test**
  - 500,000 sampling data
  - 3 inputs variables:
    - Total Fe, H and O
    - Sampling with LHC
    - Ranges taken from long term RT simulations
  - 8 output variables:
    - The valuable output variables for a RT code:
      - Total dissolved Fe, H and O
      - Total precipitated Fe, H and O
      - pH
      - Eh

Inputs

Total Fe
Total H
Total O

Batch simulation with CORE2Dv5

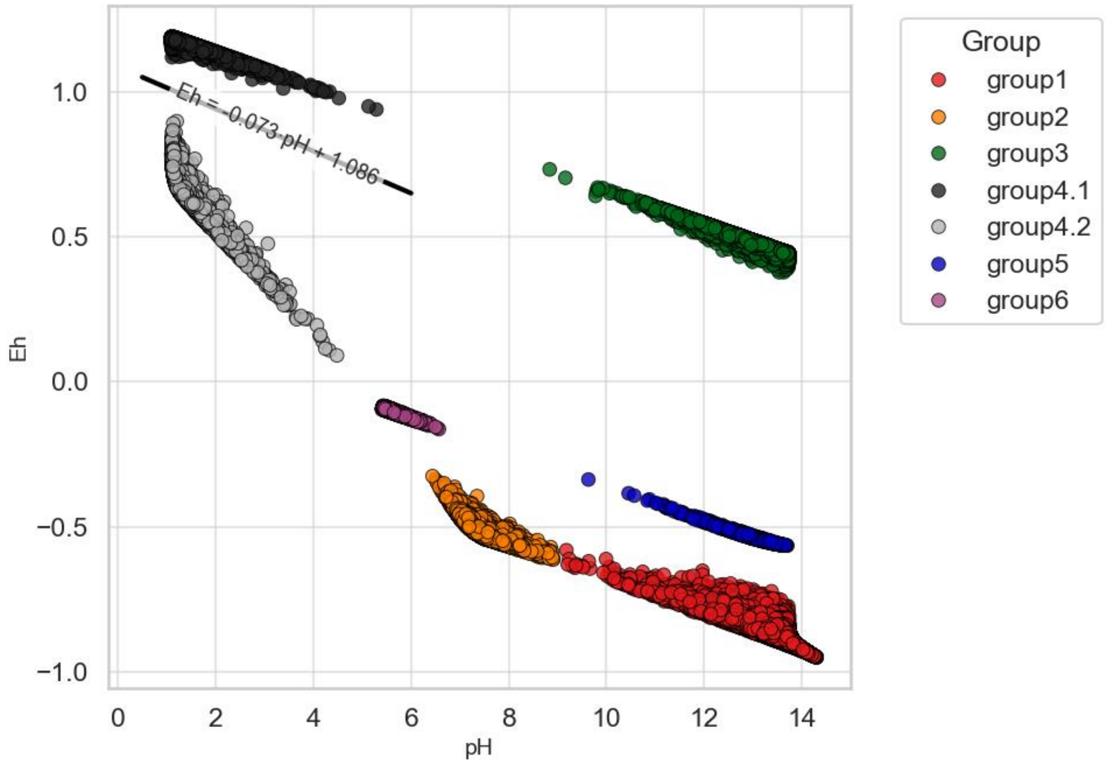
Outputs

Aqueous concentrations:
Fe^{2+}, H^{-}, O_2, OH^{-} , H_2, Fe^{3+}
Magnetite
Goethite
pH
Eh

METAMODELS FOR CORROSION

- **Grouping criteria**
  - Group 1: If Cmag > 0; Cgoethite = 0 & pH > 9
  - Group 2: If Cmag > 0; Cgoethite = 0 & pH <= 9
  - Group 3: If Cgoethite > 0; Cmag = 0 & pH > 9
  - Group 4.1: If Cgoethite > 0; Cmag = 0; pH <= 9 & [Eh > −0.12·pH + 1.16]
  - Group 4.2: If Cgoethite > 0; Cmag = 0; pH <= 9 & [Eh < −0.12·pH + 1.16]
  - Group 5: If Cmag > 0; Cgoethite > 0 & pH > 9
  - Group 6: If Cmag > 0; Cgoethite > 0 & pH <= 9

Groups	500 000 data set
1	285 774
2	53 538
3	138 576
4.1	8125
4.2	11 026
5	993
6	1968

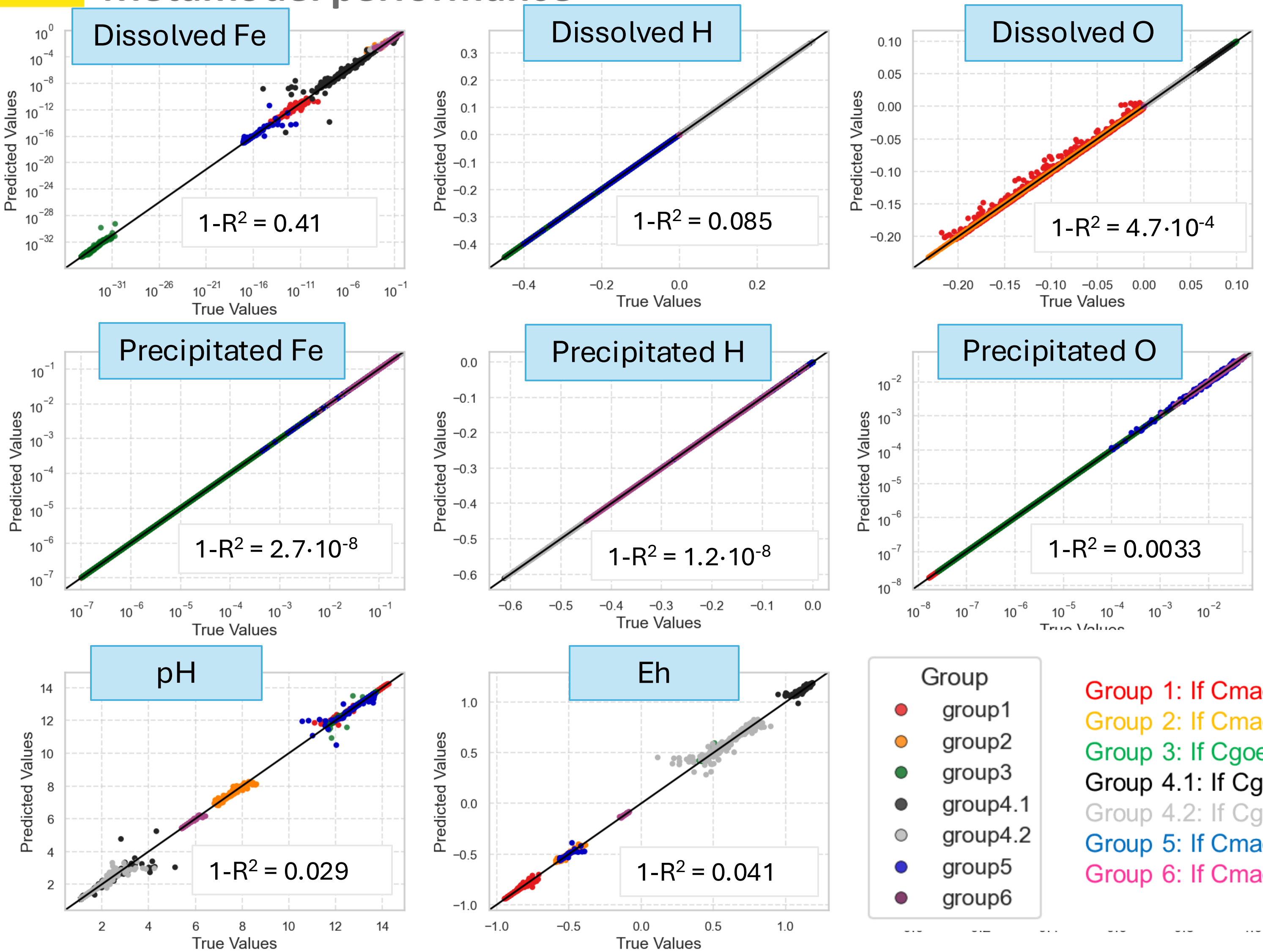


METAMODELS FOR CORROSION

- **Hybrid metamodel: RF + GP**
  - Random forest
    - 2,000 decision trees
    - Train-test split = 80 % - 20 %
    - Random shuffle before splitting
    - Accuracy of the RF model = 0.998
    - Better results were obtained in the groups with the largest number of samples
  - Gaussian Process
    - Trained with actual groups to evaluate the performance of the GP without considering RF miss groupings
    - Train-test split:
      - 80 % - 20 % for groups 5 and 6
      - 5,000 samples for the rest of the groups
    - Models trained for each variable and each group

METAMODELS FOR CORROSION

- Metamodel performance



Group 1: If Cmag > 0; Cgoethite = 0 & pH > 9
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Group 5: If Cmag > 0; Cgoethite > 0 & pH > 9
Group 6: If Cmag > 0; Cgoethite > 0 & pH <= 9

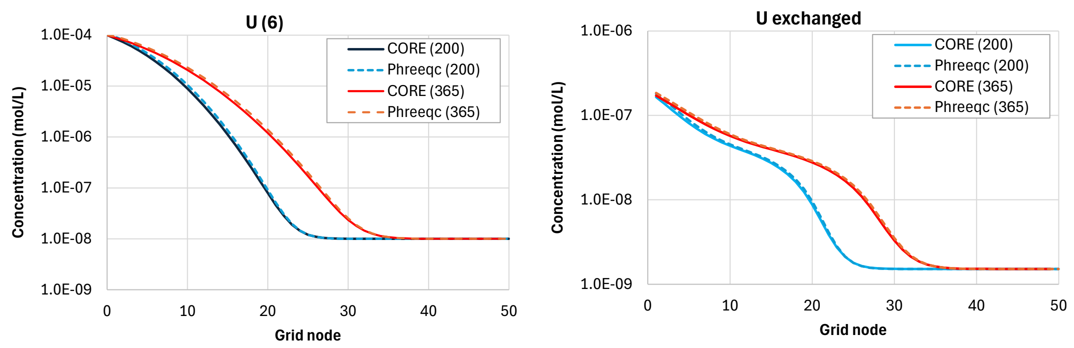
- Results:
  - Good performance metric in most groups and variables
  - Some bad performance metrics for:
    - Dissolved Fe in group 4.1 and 5
    - Dissolved O in group 1

METAMODELS FOR A URANIUM SYSTEM

- **Migration of U in a clayey host rock based on ML benchmarks in EURAD1- WP DONUT**
- **Uranium sampling cases:**
  - U exchange
    - 30,000 sampling data
    - Sampling with a real 1D RT model for diffusion and advection
    - Inputs: Total U, H, O and Na
    - Outputs: Dissolve U, O, and Na, Exchange U and Na
  - U sorption
    - 30,000 sampling data
    - Sampling with a real 1D RT model for diffusion and advection
    - Inputs: Total U, H and O
    - Outputs: Dissolved U and H, Sorbed U and H
- **Uranium metamodels:**
  - Gaussian process:
    - Train/test split = 80/20 %
    - Other parameters: like those of the corrosion case

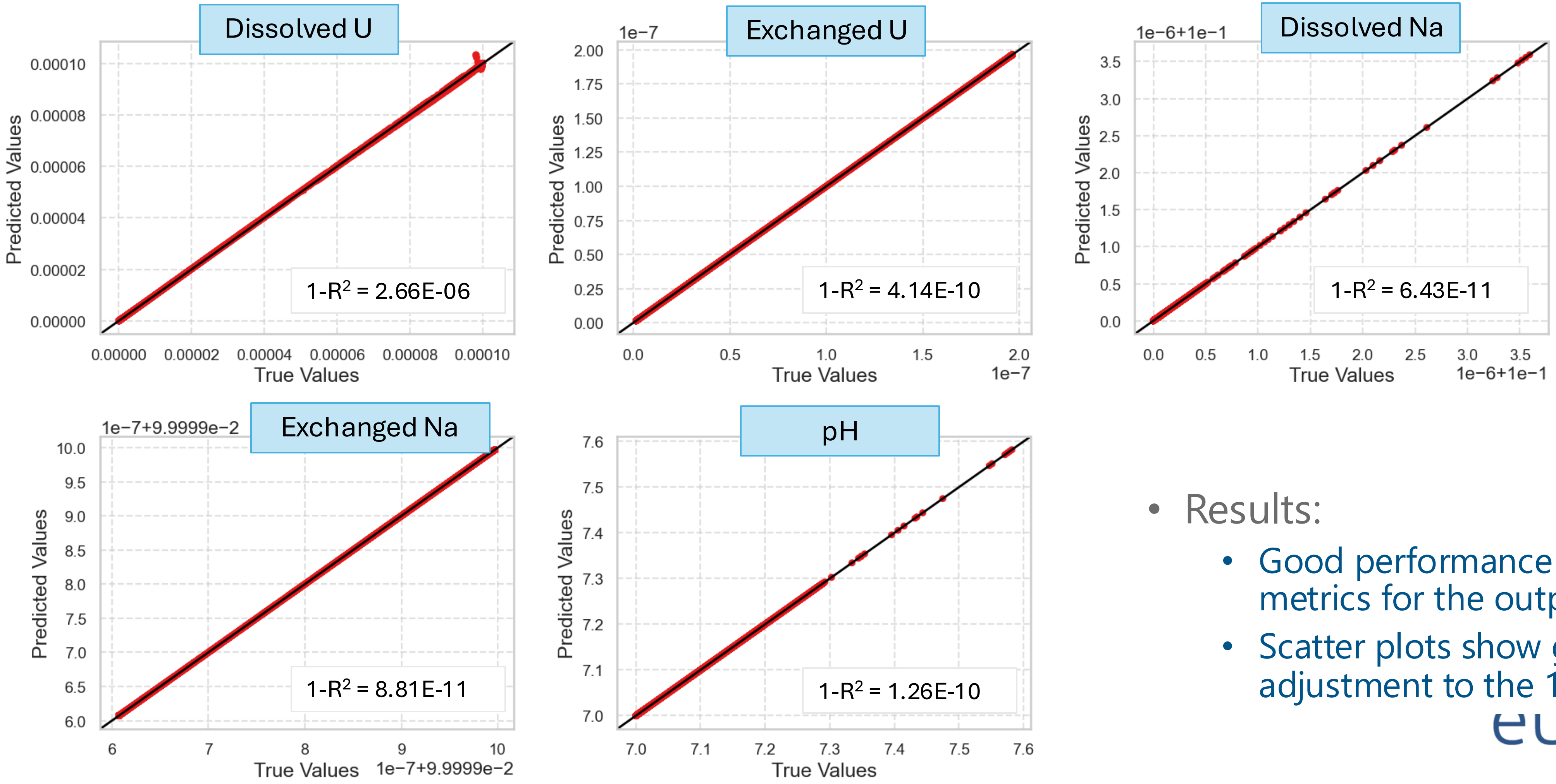
Chemistry: U-Na-Cl system
Processes: Cation Exchange – Surface Complexation
Transport: Diffusive – Advective – Advective-dispersive

Results of the 1D RT model



METAMODELS FOR A URANIUM SYSTEM

- Performance of the metamodel for Exchanged U case:



- Results:
  - Good performance metrics for the outputs
  - Scatter plots show good adjustment to the 1:1 line

CONCLUSIONS

- **The high-fidelity THC model of the FeMo corrosion test**
  - The RT model successfully reproduces the long-term laboratory observations of mineral precipitation at the iron-bentonite interface
- **Code improvements for RT simulation**
  - Progressive corrosion and magnetite passivation layer successfully capture corrosion retarding phenomena
- **Performance of the metamodels:**
  - Corrosion case
    - The hybrid Random Forest (RF) and Gaussian Process (GP) surrogate models demonstrate high accuracy (NRMSE<0.02) in predicting geochemical evolution
    - The performance is excellent for the precipitated
  - Uranium case
    - Good performance for the metamodel for most species
    - On progress the improvements of pH of the sorbed U case
  - The use of ML metamodels significantly addresses the CPU-intensive nature of geochemical solvers, enabling faster and more complex simulations

FUTURE WORK

- **Code Integration:**
  - Implementation of the trained RF and GP metamodels into the CORE2Dv5 reactive transport code
  - Metamodel performance and evaluation of the CPU time saving
- **Large-Scale Application:**
  - Global Sensitivity Analysis:
    - Perform large-scale GSA (>5,000 runs) for long-term granite repositories.
  - Dataset Expansion:
    - Expand geochemical training space for repository safety assessments.

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  - ENRESA within WP ACED of EURAD (Grant Agreement nº 847593) and WP HERMES of EURAD-2 (Grant Agreement nº 101166718)
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