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Editorial: Sorption processes in nuclear waste management: data knowledge management and new methodologies for data acquisition/prediction

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Editorial on the Research Topic
Sorption processes in nuclear waste management: data knowledge management and new methodologies for data acquisition/prediction

A fundamental approach to Nuclear Waste Repository research involves the Research Topic of experimental data in a laboratory setting, development of empirical and/or mechanistic numerical models representing those observations, and application of these models into reactive transport and performance assessment models as predictive tools for informing society of impacts and risks associated with nuclear waste repository scenarios (Stevens et al., 2020). The assimilation and interpretation of experimental data must take advantage of both new data and the rich historical data available in the literature and apply novel modeling approaches to improve predictive tools, particularly from the standpoint of uncertainty quantification, for nuclear waste repository performance assessment (Zavarin et al., 2022).

Experimental data collected in a laboratory setting is fundamentally different from large formatted sensor data that are much more amenable to "big data" approaches. New data science approaches to interrogate experimental laboratory data have been limited, in large part, by the lack of common standards and approaches to archiving these data types (i.e., findable, accessible, interoperable, reusable "FAIR" data) (Wilkinson et al., 2016). While these standards are in the process of development (e.g., DOE BER ESS-DIVE, https://ess-dive.lbl.gov/), integrating historical data (i.e., data mining) must also be part of our path forward. As these approaches are applied to sorption and other data, they will fundamentally change how predictive tools quantify impacts and risks associated with siting nuclear waste repositories.

In this series of articles, authors apply novel modeling approaches to experimental data categorized as sorption data. Traditionally, these data inform empirical (e.g., Kd) or mechanistic (e.g., surface complexation) models that are subsequently used in reactive transport models at various scales as predictive tools for assessing transport behavior. These same data and/or models can also inform the development of Reduced Order Models

(Garibay-Rodriguez et al.; Li and Zarzycki) that can be applied more efficiently to complex large scale models (e.g., performance assessment models).

Romanchuk et al. focus on the question of self-consistent Surface Complexation Model (SCM) database development by presenting a data fitting workflow that combines new and literature-derived sorption data for three radionuclides in specific oxidation states [U(VI), Eu(III), Np(V)] and the iron (III) oxide mineral goethite fit using the PHREEQC (Parkhurst and Appelo, 2013) geochemical solver linked to the MOUSE (Linge et al., 2020) optimization software with automation that relies on the Python programming language. They show that expansion of their approach to a larger set of elements [Cd(II) and Zn(II)] is relatively straightforward and suggests further expansion by adopting Linear Free Energy Relationships (LFER) across an even greater number of sorbing elements. Importantly, the adoption of LFER may be appealing but will yield additional modeling uncertainties that may be difficult to quantify or justify in nuclear waste repository performance assessment models.

The application of SCMs to predict organic compounds' behavior at solid-water interfaces is particularly complicated by the large number of organic compounds present in nuclear waste repositories and the associated near- and far-fields. Organic compounds can sorb to mineral surfaces but can also form aqueous and ternary complexes with radionuclides. As a result, developing comprehensive modeling approaches for organic compounds' reactive transport remain elusive. Szabo et al. explore this topic by examining the uptake of multiple organic compounds (degradation products of polyacrylonitrile-based polymers) to cements. They use relatively simple Langmuir models to explore the relative affinity of these organic compounds and review affinity patterns of a large number of organic compounds reported in the literature. The examination of sorption behavior across a wide number of organic compounds reveals relationships between functional group type, functional group density, and surface affinity. While this topic deserves significant additional study, it appears that exploration of sorption patterns across large number of minerals and organic compounds will lead to discoveries into new approaches for addressing the role of organic compounds in nuclear waste repository performance assessment, contaminant transport modeling, and carbon cycling in earth systems.

Hinchliff et al. examine the role of cellulosic organic compounds on Sr sorption to hydrated cements. Again, new and literature data are combined to develop a comprehensive understanding of Sr diffusivity and sorption to hydrated cements. The analysis reveals surprisingly enhanced retardation of Sr and suggests that cellulose organic degradation products impact Sr retention though the exact mechanisms have yet to be revealed. Nevertheless, the results point to the need to expand surface complexation and surrogate modeling approaches beyond simple binary radionuclide sorption models and incorporate more complex ternary organic complexes. Importantly, the increased complexity brings increased importance to the development of large FAIR sorption databases (Wilkinson et al., 2016) that capture the multidimensional sorption conditions. Development of software and workflows that can readily take advantage of the rich data sources available in the literature will greatly benefit the nuclear waste repository research community in the coming years.

An apparent roadblock in SCM database development is the apparent data paucity as it relates to development of well constrained geochemical models of relevant reactions. However, Zavarin et al. (2022) determined that the cumulative number of peer reviewed publications referencing the topic of sorption and adsorption has already reached ~1,000,000. Thus, as several authors discuss, this apparent lack of data is, in large part, due to a lack of FAIR data rather than lack of data *per se*. This lack of FAIR data has severely limited the development of self-consistent SCM databases. A notable exception is that of Dzombak and Morel (1990) who developed a mechanistic diffuse layer surface complexation database for the sorption of 22 elements to the mineral hydrous ferric oxide (HFO). A follow-on effort by Karamalidis and Dzombak (2010) developed a similar database for gibbsite. The HFO database was developed by critically evaluating 169 literature references. To this day, this database remains the most widely used database for predicting metal interaction with HFO. However, the database is static (i.e., cannot be updated) and any reactive transport models that uses this database must include the same choice of thermodynamic data (i.e., aqueous speciation constants, activity correction models) and surface complexation model (i.e., two-site diffuse layer surface complexation) employed by the original authors. A more recent data assimilation effort at Lawrence Livermore National Laboratory and collaborators at the Helmholtz Zentrum Dresden Rossendorf, has been developing a data digitization pipeline (Zavarin et al., 2022). To date, the manual digitization of data has yielded a LLNL SCIE database that includes 211 references and a total of 22,732 individual digitized data and associated metadata. Importantly, this approach to data mining is labor intensive and cannot be expanded beyond a set of targeted data (e.g., reactions specific to the nuclear waste performance assessment needs). Adoption of new approaches to data assimilation are clearly needed.

New modeling approaches that take advantage of FAIR database development efforts are also needed. Garibay-Rodriguez et al. describe a computational framework for radionuclide migration assessment in clay rocks that is based on OpenGeoSys6 which is linked to a number of geochemical solvers (PHREEQC, GEMS, etc.) and can be applied to both traditional surface complexation/ion exchange models as well as surrogate models. For portability, the modeling framework is built around the JupyterLab Python framework that guarantees that all the source code and its dependencies can be executed efficiently and reliably, independent of any computing environment. Importantly, the surrogate models (aka look-up tables) are explicitly tied to the mechanistic surface complexation/ion exchange models. Test cases suggest that surrogate models can yield significant computational speed-up while maintaining a similar level of precision in model output. Garibay-Rodriguez et al. argue that open source software approaches are key to continuing software development and longterm improvements in nuclear waste performance assessment models.

Li and Zarzycki describe a computational pipeline to generate synthetic SCM data and discuss approaches to transform this dataset into AI-learnable input for use in surrogate models. In their approach, available surface complexation model databases are used to produce high density sorption data across a very wide range of conditions. The generated synthetic datasets can more readily take advantage of AI algorithms that can be used in surrogate model development. The approach is tested for determination of the electrostatic properties of a prototypical oxide/electrolyte interface using the triple-layer surface complexation modeling construct. The

authors provide a theoretical framework for developing surrogate models using AI and based on developing synthetic sorption data from traditional surface complexation databases. While these surrogate approaches can yield computational efficiencies that may benefit performance assessment modeling efforts, it should be noted that applicability of surrogate models is often limited by the range of data available for model training.

Nuclear Waste Repository reactive transport and performance assessment modeling teams have a number of opportunities to apply modern processes, workflows, and machine learning in predictive tools for informing society of impacts and risks associated with nuclear waste repository scenarios. Both the assimilation and interpretation of experimental data can harness ML to improve predictive tools, particularly from the standpoint of uncertainty quantification. While a number of challenges are still present (e.g., automation of various component of the data assimilation and interpretation in the data stream), these approaches will provide a robust and nimble framework for evaluating nuclear waste repository performance that is adaptable to the unique timescales that repository performance, oversight, and monitoring will require. However, scientists, regulators, and society will need to have confidence that these novel modeling approaches can effectively simulate nuclear waste repository performance before these approaches can be widely accepted.

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Conflict of interest

Author DG was employed by Amphos 21.

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