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M4SF-22LL010302062-NEA-TDB and Sorption DataBase Development

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Contents

1. Introduction

This progress report (Level 4 Milestone Number M4SF-22LL010302062) summarizes research conducted at Lawrence Livermore National Laboratory (LLNL) within the Crystalline International Collaborations Activity Number SF-22LL01030206. The activity is focused on our long-term commitment of engaging our partners in international nuclear waste repository research. This includes participation in the Nuclear Energy Agency Thermochemical Database (NEA-TDB) Project (Cindy Atkins-Duffin, Mavrik Zavarin) and development of methodologies for integrating US and international thermodynamic databases for use in SFWST Generic Disposal System Assessment (GDSA) efforts.

A continuing focus for FY22 efforts has been to support the US participation in the NEA-TDB effort and developing mechanisms for integration of NEA-TDB thermochemical data (new electronic database made available in 2018) with LLNL's thermodynamic databases that support the SFWST GDSA activities. This effort is coordinated with the Argillite work package database development efforts. The goal is to provide a downloadable database that will be hosted on LLNL's thermodynamics website which incorporates NEA-TDB data into the LLNL SUPCRTNE database where appropriate. Updating thermodynamic databases and enhancement of our database integration code to ensure that SFWST GDSA efforts are based on current and internationally accepted thermodynamic data are a key focus.

As part of our international activities, we also continued our effort to integrate the JAEA Kd data into the L-SCIE database framework. In FY21, we compiled >500 lines of python code that can be used to export JAEA single mineral Kd data into a format that is compatible with the L-SCIE (Zavarin et al., 2022) database (~17,000 data). FY22 efforts focused on code debugging, data quality evaluation, and integrating these data into the L-SCIE. In addition, we began discussing data assimilation efforts from nuclear waste program databases in Germany and elsewhere. The overall goal is to produce a FAIR (Wilkinson et al., 2016) open source database that can be shared and integrated with multiple nuclear waste programs internationally. As part of this effort, we plan to hold a small meeting to discuss implementation partnerships between national and international organizations in late FY22.

2. Nuclear Energy Agency Thermochemical Database Program

2.1 History of Project

The Nuclear Energy Agency (NEA) Thermochemical Database Program (TDB) was conceived of and initiated with the goal to 1) make available a comprehensive, internally consistent, internationally recognized database of selected chemical elements; 2) meet the specialized modeling requirements for safety assessments of radioactive waste and; 3) prioritize the critical review of relevant data for inorganic compounds and complexes containing actinides. Data from other elements present in radioactive waste are also critically reviewed as well as compounds and complexes of the previously considered elements with selected organic ligands.

The objective of the Program is to produce a database that contains data for all the elements of interest in radioactive waste disposal systems; document why and how the data are selected; give recommendations based on original experimental data, rather than compilation and estimates; document the sources of experimental data; provide internally consistent thermodynamic parameters, and treat solids and aqueous species of the elements of interest for nuclear storage performance assessment calculations.

The qualification of existing data is conducted using documented Guidelines which include several components. A Technical Review is conducted by subject matter experts who critically review experimentally determined literature data; reanalyze the data as necessary; and select data for inclusion in the database. Upon completion of the Technical Review, a Peer Review is undertaken. A second, independent panel of reviewers ensure that the technical reviewers followed the review Guidelines. A Comment Resolution component ensures that the Technical Reviewers address the comments made by the Peer Reviewers. At this time, the volume is readied for final publication. Distribution of the Reviews is via the NEA TDB website.

2.2 History of Phases and New Phase 6 Efforts

Phase I of the NEA-TDB program was conducted between 1984 and 1998. Initial review volumes included Uranium, Americium, Technetium, Neptunium/Plutonium.

Phase II of the NEA-TDB program updated all the actinide volumes. The update is contained in a single volume. Added in this phase were Nickel, Selenium, Zirconium, and the compounds and complexes of the reviewed elements with selected organic ligands - EDTA, ISA, oxalate, and citrate. The Program also conducted a workshop, “The Use of Thermodynamics Databases in Performance Assessment.” The phase ran from 1998-2003.

Phase III of the NEA-TDB program saw the introduction of a second product, State-of-the-Art reports. The first such report covered solid solutions of interest to nuclear waste management. Thorium, Iron (part I), and Tin volumes were added to the published collection.

Phase IV of the NEA-TDB program, conducted from 2008-2014, started the second portion of the Iron review; review of Molybdenum, and a review of Auxiliary Data which includes species and compounds necessary to describe aqueous chemistry of Aluminum and Silicon, data on inorganic species and compounds of elements such as Iodine, Boron, Magnesium, Calcium, Strontium, and Barium.

Phase V of the NEA-TDB program, conducted from 2014-2019 took on a second update to actinide and fission product volumes, and two State-of-the-Art reviews - Cement

Minerals and High Ionic Strength Aqueous Systems. The design and development of a new TDB electronic database that is compatible with PHREEQC was undertaken.

Phases VI of the NEA-TDB program was officially started in February 2019 with all participating member parties having signed the Framework Agreement. The First Meeting of the Management Board (MB) and the Executive Group (EG) were held at the NEA in Paris February 19-20, 2019. Lena Evins (Sweden-SKB) was elected Chair of the MB and Stephane Brassinnes (ONDRAF/NIRAS- Belgium) Vice Chair. Canada and the Netherlands joined the Programme for this Phase. Elected to the Executive Group were Chair Marcus Altmaier (INE-Germany), Cindy Atkins-Duffin (DOE/LLNL-USA), Benoit Made (ANDRA-France), Pascal Reiller (CEA Saclay – France), and Kastriot Spahiu (SKB (retired)-Sweden. In process projects from Phase V were brought forward to Phase 6.

The current status of the Phase 6 program and the associated reviews are:

- **Iron (Part II) Volume** – Published online (NEA TDB website) in January, 2020.
- **2nd Update of the Actinides Volume** – Published online (NEA TDB website) in October, 2020
- **Ancillary Data Volume** – The volume is in the final stages of editing and production, expected publication (NEA TDB website) by the end of 2022. Carlos Jove-Colon (USA) served as one of the peer reviewers.
- **Molybdenum Data Volume** – This review continues to struggle with delivery of information from the team to the NEA and to other teammates. Several rescoping and personnel assignments have been put into place. The EG has taken a more active management role in this project. In preparation for peer review the first internally reviewed draft of the volume is expected to be delivered to the NEA by the end of July 2022. Final publication of the volume is anticipated in the first quarter of 2024.
- **Cements State of the Art Report** – Considerable discussions were conducted by the TDB MB and EG regarding the pace of this review. The NEA Project Coordinator and the EG liaison (Cindy Atkins-Duffin, USA) convened quarterly meetings with the review team to monitor the review teams progress to their schedule. In preparation for peer review the first internally reviewed draft of the volume is expected to be delivered to the NEA by the end of July 2022. Final publication of the volume is anticipated in the first quarter of 2024.
- **High Ionic Strength Solutions State of the Art Report** – Progress continues to be slow on this project. The next deliverable, first single author drafts and internal reviews of the text are due July 2022. The current planned publication (NEA TDB website) is in the first quarter of 2024.
- **Organics Update** – The initiation report has been completed and the Management Board accept the report as presented, paid, and closed out the reviewer contracts. The Management Board has prioritized the completion of the existing volumes higher than the start of new activities. While recognizing the importance of this Update, the decision was made to defer the start of this effort to a possible Phase 7 of the TDB Programme.

- **Lanthanides Volume** – This activity held its Kick-off meeting in late 2021. Work on this volume continues with anticipated publication date of the end of 2025.
- **High Temperature State of the Art Report** – The initiation report has been completed. The Management Board requested two changes be made to the report (removal of specific cost estimates and addition of notional project timeline) at which time the report will be accepted. The Management Board has prioritized the completion of the existing volumes higher than the start of new activities. While recognizing the importance of this Update, the decision was made to defer the start of this effort to a possible Phase 7 of the TDB Programme.
- **TDB course** – The 6th edition of the course is being organized as a Webinar for November 15-16, 2022. All registration and agenda materials are available online. As of end of June 2022 50 participants have registered. The 7th edition of the course is being planned as an in-person event to be held in conjunction with Migration 2023, Nantes, France.
- **TDB Electronic Database** – Currently only maintenance activities such as the populations of the eTDB with the new selected values from the Ancillary and Molybdenum volumes are being undertaken. All available NEA staff resources have been focused on the management, editing, and publication of the review volumes.

Meetings continue to be held in the virtual format. It is anticipated that the annual Management Board meeting in November 2022 will be held in person in Paris.

The current U.S. representative to the NEA TDB Management Board, Cynthia Atkins-Duffin, has announced her intentions to retire from LLNL. Mavrik Zavarin, also from LLNL, will replace her on the Management Board. The Management Board will fill her vacancy on the Executive Group.

3. Japanese Atomic Energy Agency Kd Database Assimilation Effort

In an effort to build a FAIR(Wilkinson et al., 2016) open source database of raw sorption data for use in nuclear waste performance assessment models, we reached out to our colleagues at the Japanese Atomic Energy Agency and began the process of assimilating JAEA's large database of sorption data into the L-SCIE(Zavarin et al., 2022) database in FY21. The JAEA Kd database is a web based open source database that contains nearly 70,000 Kd data (https://migrationdb.jaea.go.jp/nmdb/db/sdb/search_1.jsp). The database includes information on both rock and single mineral sorption data. The database also spans a wide range of radionuclides relevant to the nuclear waste programs. However, a number of issues needed to be resolved to assimilate these data into the LLNL SCIE database for use in surface complexation/ion exchange database (and ML) development being pursued at LLNL. Key issues to be resolve included the following:

- Conversion of electrolyte composition into units compatible with the L-SCIE (mol/L)
- Conversion of gas composition alphanumeric strings into gas fugacity values
- Conversion of various alphanumeric strings (value ranges, "<" and ">" values, etc., into real numbers

- Conversion of Kd data into aqueous and sorbed radionuclide concentration.

These issues were largely resolved in late FY21. In this first effort to assimilate JAEA data, nearly 17,000 single mineral data were converted into the L-SCIE format. In FY22, we developed a python code to integrate the JAEA single mineral data with the L-SCIE database and workflow. This has led to a combined L-SCIE/JAEA dataset that contains 44,000 sorption data. This is by far the largest accumulation of FAIR sorption data assembled in a single database and presents an important step in the development of data-driven workflows to simulate the sorption phenomenon that is critical to the prediction of radionuclide migration in reactive transport and performance assessment models. We illustrate the power of this database in the Section 4 of this report.

In FY22, we also began developing additional international collaborations around the theme of database interoperability and collaboration. We have continued our discussions with the RES^{3}T developers at the Helmholtz Zentrum Dresden Rossendorf. This collaboration led to a joint publication of our surface complexation database and workflow named L-SCIE (Zavarin et al., 2022). The RES^{3}T group was recently awarded a grant to pursue sorption database development for Germany and we are in close contact to ensure that both the US and German databases are interoperable. This effort is being pursued together with partners at the Paul Scherrer Institute in Switzerland which holds a large collection of clay sorption data. These data are being assembled into a FAIR database and we are working to ensure that these data can be shared between the various international nuclear waste partners as well. We have also been working with David Garcia at AMPHOS^{21}, a consulting company based in Spain that was contracted to develop a sorption database for Stephane Brassinnes of the Belgian nuclear waste program. We are working closely with this group to ensure database compatibility between the L-SCIE and Ondraf-Niras databases. Finally, we are in contact with Dr. Zimeng Wang at Fudan University (China). Dr. Wang has been involved in sorption data digitization and modeling efforts and has been interested in collaborating to ensure interoperability across various international sorption database efforts.

In late FY22, we collaborated with Dr. Garcia to develop a special research topic in the Frontiers in Nuclear Engineering journal entitled "Sorption processes in nuclear waste management: Data knowledge management and new methodologies for data acquisition/prediction". We are currently gathering manuscript contributions and the full research topic will be published in FY23. We anticipate that a number of new international data streams will become available in FY23 and we will continue to pursue international collaborations that promote interoperable design of databases and the principles of Open Science and FAIR data.

4. Workflow development for community potentiometric titration data modeling and database development

4.1 Introduction

Since the surface charge of the mineral affects to sorption affinity between adsorbate and adsorbent, estimating the protolysis of mineral surface is crucial for surface complexation models (SCM). For this reason, surface acidity constants of minerals, which describe degree of the protonation and deprotonation of surface functional groups, are essential input parameter of SCMs and the selection of proper values for the constants is important in terms of reliable modeling. In various studies, however, different surface acidity constants have been utilized even when the type of SCM and mineral used in their studies are identical (see Table 1). This fact implies that selecting a single representative value of surface acidity constant involves ambiguity and it is necessary to produce a universal surface acidity constant. Our task aims at developing surface titration modeling workflow based on community potentiometric titration data to produce ‘global’ surface acidity constants. This approach is now possible given the extensive FAIR data that are available in the L-SCIE database. Below, we illustrate this approach using ferrihydrite as our model mineral system.

Table 1. Surface acidity constants used in various studies using diffuse double layer (DDL) model. All data taken from RES^{3}T database (Brendler et al., 2003; Dresden-Rossendorf, 2013).

	1st surface acidity constant (pK_1)	**2nd surface acidity constant (pK_2)**	**Associated surface complexation reaction**	**Reference**
1	6.09	7.38	»Fe-OH + Ni-EDTA^{2-} + H^+ ↔ »Fe-Ni-EDTA$^-$ + H_2O	(Nowack et al., 1996)
2	6.51	9.13	»Fe(s)-OH + UO_2^{2+} ↔ »Fe(s)-O-UO_2^+ + H^+	(Arnold et al., 1998)
3	7	9.2	»Fe(w)-OH + Cd^{2+} ↔ »Fe(w)-O-Cd^+ + H^+	(Nomaan et al., 2021)
4	7.01	7.86	»Fe-OH + CrO_4^{2-} + H^+ ↔ »Fe-CrO_4^- + H_2O	(Veselská et al., 2016)
5	7.29	8.93	»Fe(w)-OH + Cd^{2+} ↔ »Fe(w)-O-Cd^+ + H^+	(Eibl et al., 2019)
6	7.5	10.2	»Fe-OH + Co^{2+} ↔ »Fe-O-Co^+ + H^+	(Landry et al., 2009)

4.2 Progress to date

4.2.1 Development of surface titration modeling framework

As part of this effort, a community data driven surface complexation modeling workflow has been developed. The workflow begins with a database of community titration/sorption data, i.e., L-SCIE (Zavarin et al., 2022), followed by a surface titration modeling workflow, which is then followed by surface complexation modeling workflow (Figure 1). The L-SCIE database provides input data for both surface titration modeling (L-ASTM, LLNL Automated Surface Titration Model) and surface complexation modeling (L-ASCM, LLNL Automated Surface Complexation Model). The surface acidity constants produced by L-ASTM are used as input parameters for the surface complexation model (L-ASCM).

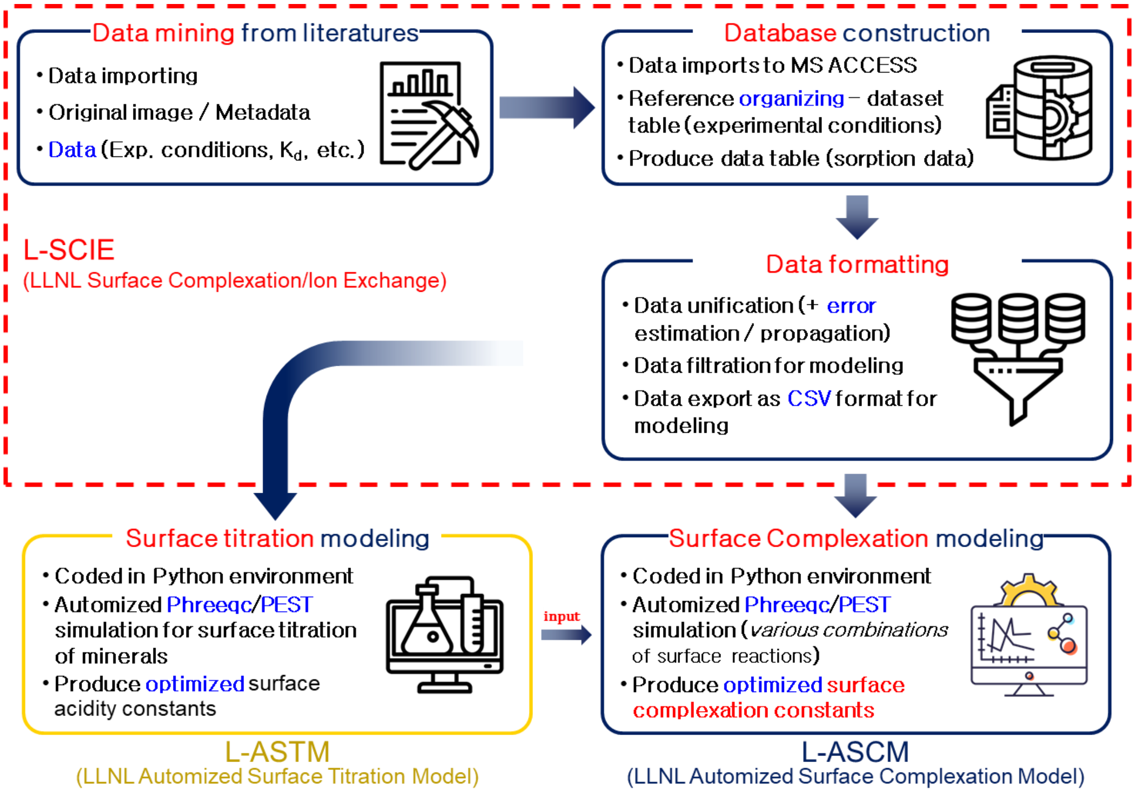

Figure 1, Workflow of community data driven surface complexation modeling framework

Data processing and workflow associated with L-ASTM was written in Python and the code was integrated with PHREEQC (Parkhurst and Appelo, 2013) and PEST (Doherty, 2018) for data fitting. The details of the modeling workflow of L-ASTM are shown in Figure 2. First, the code imports potentiometric titration data extracted from the L-SCIE database and categorizes the data by dataset. For example, single reference may include several experimental datasets conducted under various experimental conditions, e.g., ionic strength. In the L-SCIE database, there are 40 surface titration datasets for ferrihydrite which are obtained from 12 references (See Table 2). For each of these datasets, the L-ASTM code generates the relevant PHREEQC/PEST input files and runs a separate PHREEQC/PEST data fitting. Simulation results are evaluated by using various criteria, e.g., goodness of fit, and the code selects acceptable datasets according to the quality of fit obtained for each dataset (defined as "qualified" datasets). Average surface acidity constants of "qualified" datasets are then calculated. Finally, the code runs a global PHREEQC/PEST simulation using "qualified" datasets and produces optimized surface acidity constants.

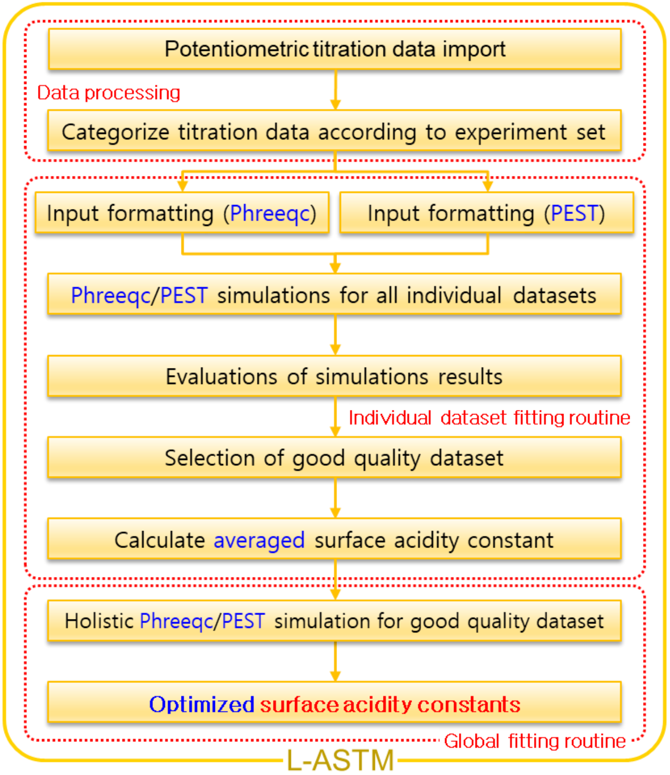

Figure 2. Surface titration modeling workflow associated with the L-ASTM code.

4.2.2 Case study of surface titration modeling for ferrihydrite

Here, we provide a case study of surface titration modeling for ferrihydrite to demonstrate the newly developed L-ASTM code. The current case study was developed based on the community potentiometric titration data (1,136 datapoints) extracted from 12 references (Antelo et al., 2010; Davis, 1978; Dyer, 2003; Ghoneimy et al., 1997; Hofmann et al., 2005; Hsi and Langmuir, 1985; Kanungo and Mahapatra, 1989; Moon and Peacock, 2013; Nagata and Fukushi, 2010; Nagata et al., 2009; Pivovarov, 2009; Trivedi and Axe, 2001) and employed a diffuse double layer (DDL) model (Table 2). In the first step, PHREEQC/PEST optimization is performed for each dataset individually, totaling 40 runs in this case. The fit quality of each dataset was evaluated based on weighted Pearson correlation coefficient (R value) and standard deviation (SD) of surface acidity constants. If the R value is higher than 0.9 and SDs of all constants are lower than 1.0, it is presumed that the fit of the dataset is good and the dataset is deemed “qualified”. The average surface acidity constants of “qualified” datasets are 6.81 and -8.77 for pK_1 and pK_2, respectively. The global fit to the surface titration data was also conducted for these “qualified” datasets. To complete this modeling effort, the code consolidates all “qualified” datasets and PHREEQC/PEST produces optimized global surface acidity constants (Figure 3). The obtained R value of the global data fitting exercise was 0.9234 and calculated surface acidity constants were 6.76 and -8.84 for pK_1 and pK_2, respectively, which are almost identical to averaged values.

Table 2 Surface titration PHREEQC/PEST data fitting for ferrihydrite

Total number of references	**Total number of datasets**		**Total number of datapoints**
12	40		1136
Surface complexation reactions	**Initial log$_{10}$K**	**Averaged log$_{10}$K**	**Fitted log$_{10}$K (global)**
»Fe-OH + H^+↔ »Fe-OH_2^+	7.29	6.81	6.76
»Fe-OH ↔ »Fe-O^- + H^+	-8.93	-8.77	-8.84

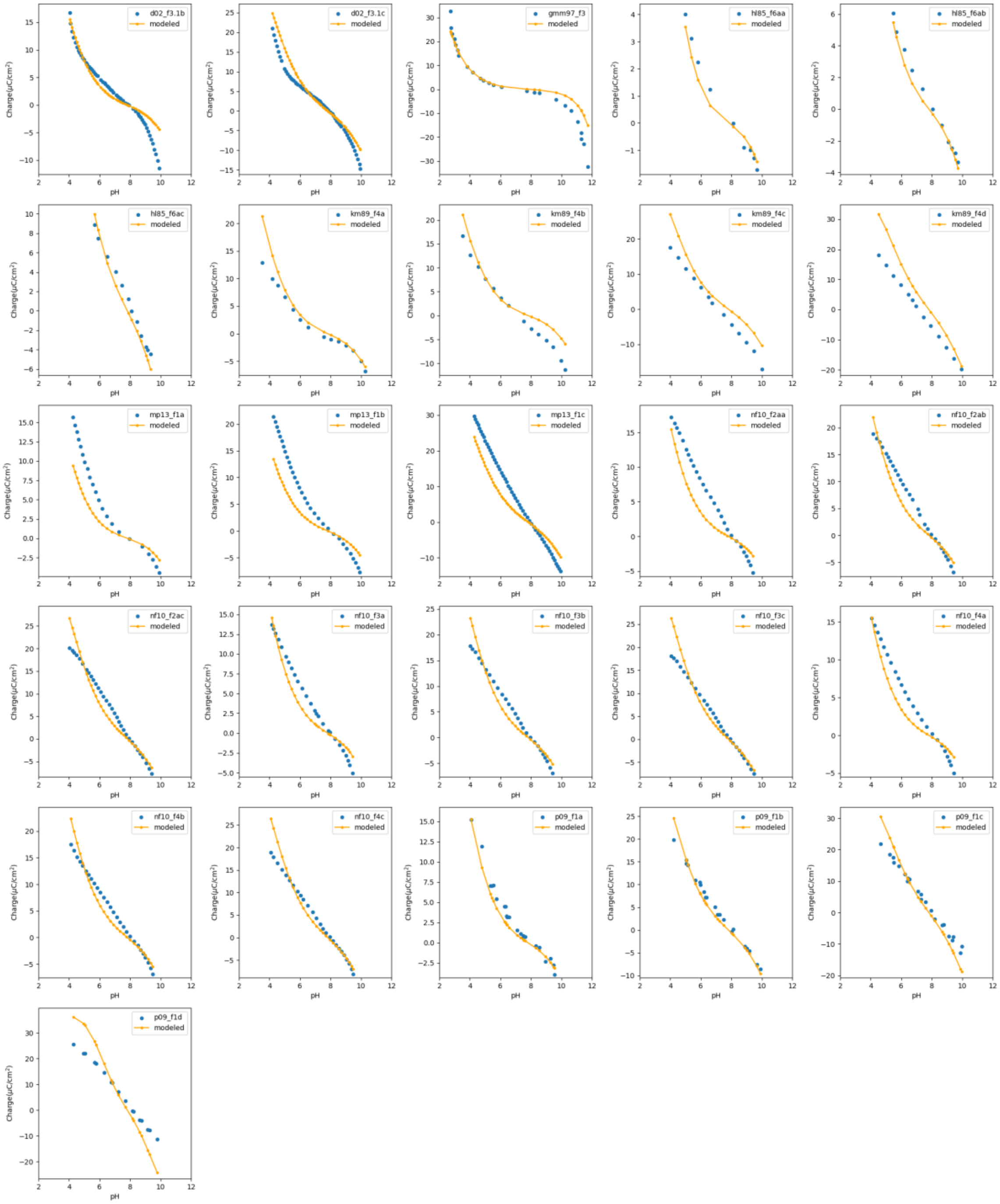

Figure 3. Simulation results for surface titration of ferrihydrite. Datapoints colored in blue represent experimental data and orange solid lines represent global data fitting results using L-ASTM.

4.3 Next steps

The model developed through the present task showed its ability to simulate the potentiometric titration data for a mineral surface. However, several additional workflow improvement efforts are needed. Most importantly, surface site density (SSD) is a unique feature of an adsorbent and an essential input parameter of the surface titration data fitting workflow. The current model used an arbitrary SSD value, 2.31 sites/nm^2 and the fitting acidity constants are very sensitive to the choice of SSD. In the future, we plan to optimize the SSDs based on a crystallographic approach (Eibl et al., 2019; Neumann et al., 2021). In addition, sensitivity analysis for the SSD can be considered to understand the effect of SSD on surface acidity constants estimations. Importantly, the automated workflow described herein also makes testing of various SCM constructs (e.g. non-electrostatic, diffuse layer, triple layer, etc.) and parameter sensitivity analyses possible even when optimizing 40 individual datasets that contain over 1000 datapoints and a single mineral surface (i.e. ferrihydrite in our case study described herein).

5. Planned FY23 Efforts

A continuing focus for FY23 efforts will be to support the US participation in the NEA-TDB effort and developing mechanisms for integration of NEA-TDB thermochemical data with LLNL's SUPCRTNE thermodynamic database that supports the SFWST GDSA activities. This effort is coordinated with the Argillite work package SUPCRTNE database development efforts. The goal is to provide a downloadable database that will be hosted on LLNL's thermodynamics website which incorporates NEA-TDB data into the LLNL database where appropriate. Updating the LLNL SUPCRTNE thermodynamic database with NEA-TDB data and enhancement of our database integration code to ensure that SFWST GDSA efforts are based on current and internationally accepted thermodynamic data are a key focus in the next fiscal year.

As part of our international activities, we will continue our effort to integrate international sorption databases into the L-SCIE database. FY23 efforts will focus on ensuring interoperable database development across multiple international database development activities. We plan to publish a complete analysis and workflow for modeling community surface titration data and illustrate the approach using a series of iron oxide minerals as examples (hydrous ferric oxide, goethite, hematite, and magnetite). The overall goal is to produce an open source database that can be shared and integrated with multiple nuclear waste programs internationally and harness modern datascience workflows and algorithms to incorporate these new approaches into reactive transport and performance assessment models.

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