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Enabling data-driven process dynamic modeling for extractive leaching and chemical precipitation

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ABSTRACT

To address the limitations of static models and gain insight into the processes of extractive leaching and chemical precipitation, a data-driven dynamic modeling strategy is proposed using a Lithium-ion battery recycling case study. The data correlations among pH, temperature, redox potential, conductivity and system state are investigated. Predictive models are then developed to describe the system state online and are employed as surrogate models for time-intensive offline chemical analyses. This enables further process optimization, such as time-saving measures and improved process efficiency through dynamic parameter studies. The proposed strategy serves as a guideline for dynamic modeling and integrates big data methodologies into chemical engineering.

1. Introduction

In the fields of chemical engineering, machine learning is widely applied in materials design and process optimization (Liu et al., 2020; Mobarak et al., 2023; Vasudevan et al., 2021). To reduce unnecessary laboratory analyses, Kida et al. (2024, 2023) applied machine learning methods to predict pollutant emissions during the leaching of contaminants from microplastics. Mehrpour et al. (2023) utilized Artificial Neural Networks (ANNs) to predict the optimum pH for heavy metal removal from wastewater through chemical precipitation. In the water quality analysis, García-Alba et al. (2019) and Ahmed et al. (2019) demonstrated the viability of employing ANNs to identify non-linear and complex relationships between input and output data.

Most publications related to machine learning in minerals processing center around machine vision (McCoy and Auret, 2019). In extractive metallurgy, Ma et al. (2018) created ANN models to predict the Total Rare Earth Elements extraction efficiency, which augments the predictability and controllability of the recycling processes (Estay et al., 2023).

In battery recycling, leaching and chemical precipitation are essential methods for separating valuable metal elements from the cathode material. Most studies focus on improving extraction efficiency by modifying static conditions, such as solvent type and concentration, solid-liquid ratio, temperature, and residence time. In precipitation, the research centers on the type and concentration of the precipitation agent, temperature and separation pH. However, the dissolution and precipitation rates of metal elements differ due to selectivity and their chemical characteristics. Dynamic models can provide insights into the extraction status and support process optimization such as shortening the extraction time. Furthermore, research into linearity and non-linearity is necessary for model selection, which is not covered in the above publications.

Fig. 1 illustrates a conceptual model that describes the data relationships in extractive leaching and chemical precipitation. In machine learning and pattern recognition, a feature is an individually measurable property or characteristic of a phenomenon (Bishop, 2006). The phenomena are described by state indicators. Both features and state indicator are determined by the state of a chemical system, which includes composition, structure, temperature T, pressure P, and pre-history data. For leaching and chemical precipitation, the most important features include pH, temperature change, redox potential and conductivity. The reaction quotient Q_r quantifies the relative amounts of products and reactants. It correlates with features and can be derived further to support the process optimization.

This paper introduces a strategy designed to enable process dynamic modeling with online sensor measurements and chemical analysis data in extractive leaching and chemical precipitation processes.

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2. Material and experimental procedure

The material utilized in this study comprises vacuum thermally treated black mass from Lithium-ion batteries. It consists of 4.2 % lithium, 14.2 % nickel, 9.5 % manganese, 10.8 % cobalt, 4.6 % aluminium, 1.2 % iron, and 2.2 % copper. The black mass was first leached with 1 molar sulfuric acid under 1:10 solid-liquid ratio for one hour, followed by precipitation with sodium hydroxide (Wang and Friedrich, 2015). Valuable metal elements from the black mass were first dissolved in the acidic solution and then separated at different pH levels as mixed hydroxide precipitates. Eight leaching and eight precipitation experiments were conducted at varying temperatures, ranging from room temperature to 75 °C. Fig. 2 illustrates the experimental setup.

3. Data-driven modeling strategy

The creation of process dynamic modeling involves multiple stages. Fig. 3 provides a visualization of the proposed data-driven modeling strategy. The process begins with data acquisition, followed by the correlation study between sensor data and system state. The data are interpolated for the purpose of non-linear modeling. Predictive models are then created using both linear regression and ANNs to predict the system state based on real-time sensor data.

Various features of a chemical reactive system, such as the activity of hydrogen ions, the average energy of the particles, the oxidation or reduction tendency of a solution, and the ability to conduct electric current, can be described by physical measurements of pH, temperature, redox potential, and conductivity, respectively. These are the features typically considered in leaching and chemical precipitation processes and can be measured online. In this study, pH range was calibrated from 0 to 14, temperature from 0 to 100 °C, redox potential from − 500 mV to 500 mV, conductivity from 0 to 2000 mS/cm.

To monitor the progress of chemical reactions, multiple samples were taken during the experimentation. The chemical analysis of the samples was performed using ICP-OES element analysis. The reaction quotient Q_r was replaced for simplification by leaching state and precipitation state for leaching and precipitation, respectively. They were calculated using current metal content in solution divided by the initial metal content in feedstock. All data were recorded with a timestamp. Fig. 4 illustrates the leaching state of Ni along with the corresponding pH, conductivity, and redox potential data from a single experiment. There appears to be potential correlation between the chemical analyses and the sensor data.

The monotonic relationship between a single feature and system state was initially studied using Spearman's rank correlation coefficient r_s. Linear relationship was then quantified with Pearson's correlation coefficient $\rho_{X,Y}$. The calculations are based on Eqs. (1) and (2), where $\mathrm{cov}(X, Y)$ is the covariance of X and Y, σ is the standard deviation, R(X) and R(Y) are the ranks of X and Y (Myers et al., 2010).

$$r_s = \quad \rho_{R(X),R(Y)} = \mathrm{cov}(R(X), R(Y))/(\sigma_{R(X)}\sigma_{R(Y)}) \tag{1}$$

$$\rho_{X,Y} = \mathrm{cov}(X, Y)/\sigma_X\sigma_Y \tag{2}$$

According to the rule of thumb for Interpreting correlation coefficient, two parameters are considered highly correlated if the correlation coefficient exceeds 0.7 (Hinkle et al., 2003). From the correlation matrices, the monotonic and linear relationship between single features can be determined. However, linear data relationship could also exist between combinations of features and system state. To avoid the issue of multicollinearity, independent features with a Variance Inflation Factor (VIF) less than 5 can be utilized in a Multiple Linear Regression (MLR) model (James et al., 2013). The adjusted R-squared scores serves as the evaluation criterion for comparing the linear regression models with different input features.

Correlation analysis and MLR modeling indicate the potential use of redox potential in describing the precipitation state of Al at the sampling points during precipitation. However, the measurement of redox potential was not limited to these sampling points. It was continuously measured throughout the entire experiment. Theoretically, the precipitation states of Al between sampling points can also be predicted using redox potential. Furthermore, the leaching data illustrates poor linearity, particularly for Li and Mn. To facilitate the application of data-driven methods and provide benchmarks for model evaluation, system states between sampling points were interpolated based on time sequence and pH-value in leaching and precipitation, respectively.

After interpolation, the data volume increased to a level where non-linear data-driven methods could be applied. In this study, a simple Artificial Neural Network (ANN) model with a Long-Short-Term-Memory (LSTM) layer and two dense layers was applied. Linear regression models were reconstructed using interpolated data. The data were normalized using the standard scaling method to enhance the generalizability of the model's prediction capability on new datasets.

4. Model evaluation and discussion

Mean Squared Error (MSE) and Mean Absolute Error (MAE) were used to evaluate the performance of the regression models on the test set. They were calculated based on (3) and (4) with scikit-learn library,

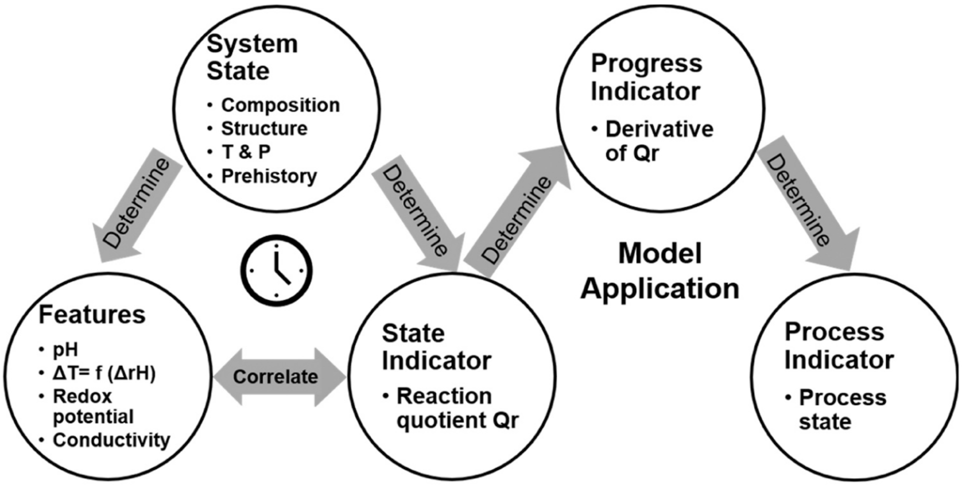

Fig. 1. Concept of modeling strategy in leaching and chemical precipitation. T for temperature; P for pressure; ΔrH for standard enthalpy of reaction change; Qr for reaction quotient.

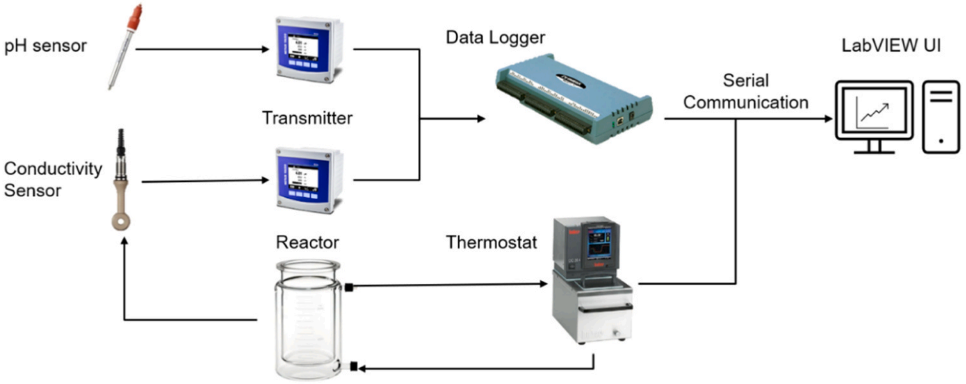

Fig. 2. Experimental setup and measurement devices.

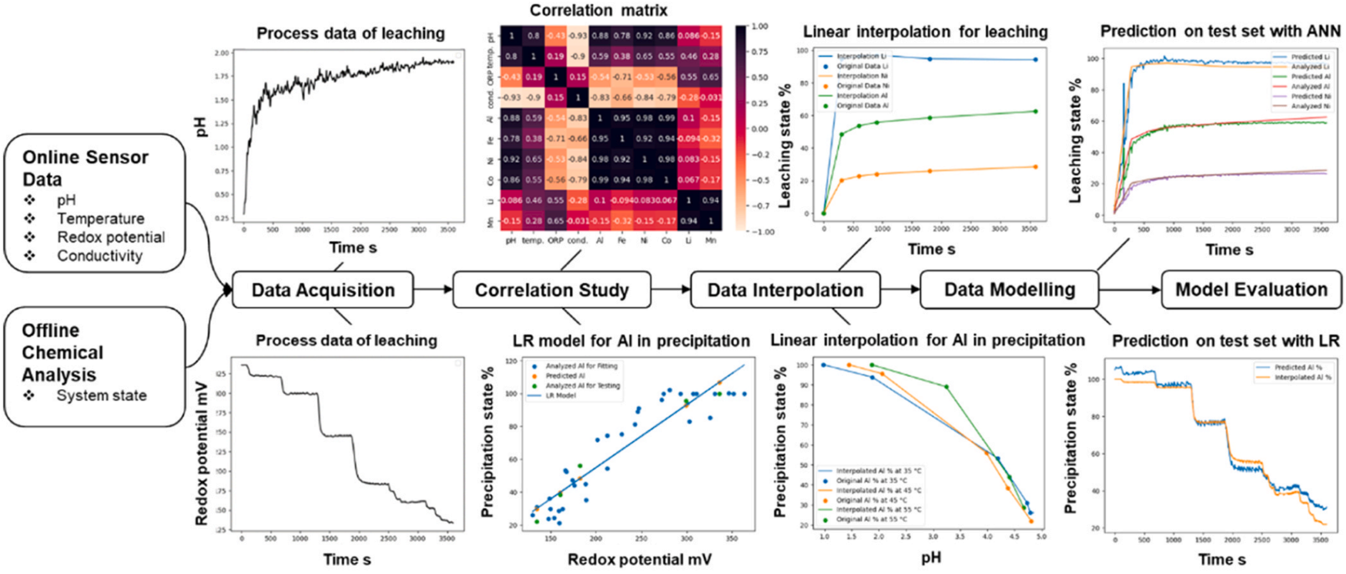

Fig. 3. Overview of the data-driven modeling strategy.

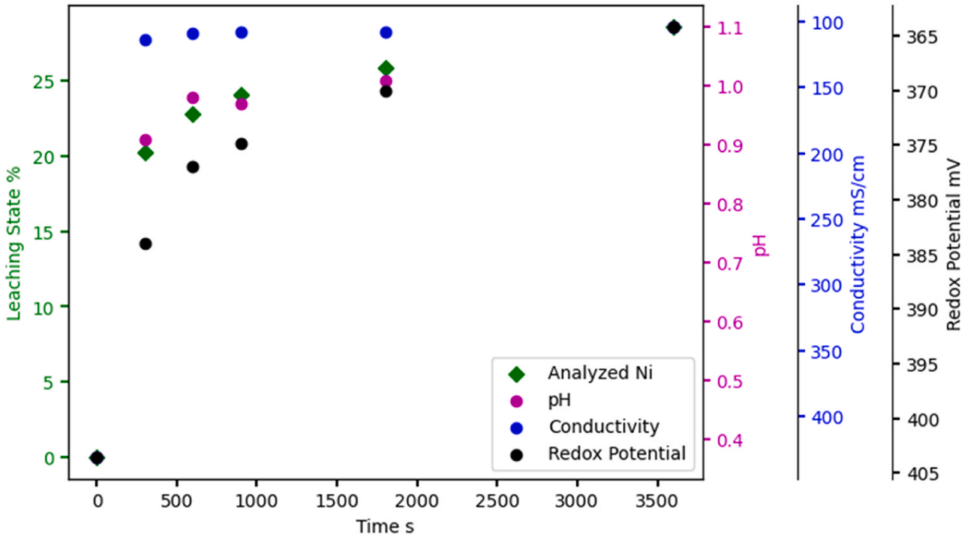

Fig. 4. Visualization of chemical analyses, pH, conductivity and redox potential.

Table 1
Evaluation and comparison of LR models and ANN models for leaching experiments.

Input features	Model	MSE_Al	MAE-Al	MSE_Fe	MAE-Fe	MSE_Ni	MAE-Ni	MSE_Co	MAE-Co	MSE_Li	MAE-Li	MSE_Mn	MAE-Mn
pH	LR	67.7	6.7	85.3	7.7	8.7	2.5	47.5	5.4	199.9	9.9	198.2	9.7
	ANN	20.1	3.8	27.8	4.6	4.9	1.9	12.9	3.0	19.7	1.8	19.3	1.7
Temperature & Conductivity	LR	42.0	3.2	40.4	3.6	13.0	2.0	30.4	2.3	86.0	4.9	85.7	4.9
	ANN	5.4	1.6	6.0	2.0	1.8	1.0	4.6	1.4	9.6	2.2	11.4	2.5

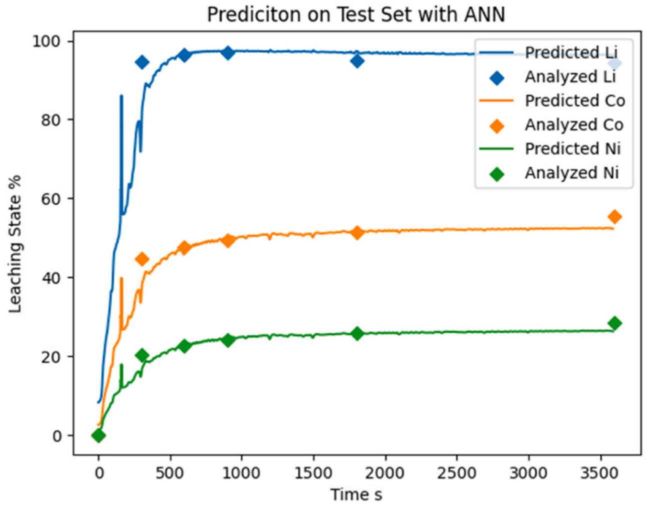

Fig. 5. Comparison ANN model predictions and chemical analyses.

where Y is the observed value and $\widehat{Y}$ is the predict value. As depicted in Table 1, ANN models offer improved leaching state predictions by accounting for potential non-linearities. Fig. 5 illustrates the leaching state predictions made by the ANN model.

$$MSE = \frac{1}{n}\sum_{i=1}^{n}(Y_i - \widehat{Y}_i)^2 \quad (3)$$

$$MAE = \frac{1}{n}\sum_{i=1}^{n}|Y_i - \widehat{Y}_i| \quad (4)$$

5. Conclusions

The physical dissolution and chemical reactions can be tracked with the support of online measurements and data-driven predictive models, offering a new perspective for possible process optimization. This work contributes to the development and verification of a strategy for process control based on the experimentally observed correlation between process parameters and performance indicators. The results demonstrate that artificial neural networks are proficient in modeling non-linear data relationships and are well-suited for leaching state prediction. This strategy serves as a general guide and is thus applicable to the dynamic modeling of other extractive leaching and chemical precipitation processes. The following steps need to be followed:

- Digitalization and data acquisition.
- Correlation study.
- Data interpolation.
- Data modeling.
- Model evaluation.

To implement the proposed data strategy in industrial processes, a robust measurement and data infrastructure must be established. Industrial data are larger and more complex than lab data, requiring additional effort in data pre-processing. Defining concrete use cases is essential as the starting point for correlation analysis. Furthermore, high-end GPUs are necessary for training data-driven models and making predictions. By integrating these models into online monitoring systems, operators can make informed decisions and gain better control over the process, ultimately enhancing the overall performance and sustainability of the extraction process.

CRediT authorship contribution statement

Bernd Friedrich: Supervision, Resources. **Tobias Kleinert:** Supervision. **Andrey Yasinskiy:** Writing – review & editing, Visualization, Methodology, Conceptualization. **Fabian Diaz:** Writing – review & editing, Supervision, Funding acquisition. **Wei Song:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Investigation, Formal analysis, Data curation, Conceptualization.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data Availability

The data presented in this study are available on request from the corresponding author.

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