A hybrid prediction model with a selectively updating strategy for iron precipitation process in zinc hydrometallurgy

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Outline

- Introduction
- Problem Formulation
- Main Results
- An Example
- Conclusion and Future Work
Introduction

- Modeling methods of industrial systems
  - Mechanism modeling method
    Physical and chemical reaction analysis of industrial systems

    **Advantage:** Clear physical meaning
    **Shortcoming:** Unmodeled dynamics in the model

  - Data-based modeling method
    Regression analysis between input and output variables

    **Advantage:** Strong applicability and generality
    **Shortcoming:** Relying too much on the quantity and quality of samples which are difficult to obtain in industrial systems
Introduction

- Hybrid modeling method

A hybrid model combines a mechanism model with a data-based model.

A set of methods for parameter identification and updating strategy for the hybrid model is developed.

Fig. 1 Framework of a hybrid model

- $x$: States
- $y$: Outputs
- $u_1, u_2$: Controls
- $\xi$: Unmodeled dynamics
Introduction

- **Modeling methods for iron precipitation by goethite**
  - A mechanism model of the iron precipitation process based on the reaction kinetics and mass balance
  - An integrated model of the iron precipitation process by combining a mechanism model with an error compensation strategy

These works have not considered the unmodeled dynamics in the mechanism model. It is necessary to deal with the unmodeled dynamics by data-based modeling!

A hybrid modeling method is proposed for the iron precipitation process by goethite in this paper.
Outline

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The aim of the process is to remove ferrous and iron ions from the zinc sulfate solution to ensure quality of zinc ingot product.
Problem Formulation

- **Reactions in iron precipitation process by goethite**

The ferrous ions are oxidized to iron ions by oxygen and iron ions are hydrolyzed to form the goethite precipitate in a zinc sulfate solution. In order to maintain the pH value, zinc oxide is added to neutralize the hydrogen ions.

- **The reaction conditions have to be strictly controlled.** If ferrous ions are oxidized and precipitated too quickly or too slowly, both the iron removal rate and the goethite precipitate quality will be poor.
Problem Formulation

- CSTR system for a single reactor

**Problem Formulation**

The oxidation rate of the ferrous ions is an important factor, which is affected by concentration of dissolved oxygen ($c_{O_2}$).

**Fig. 4 The reaction unit of the #1 reactor**

- **V**: Volume of the reactor
- **$F$**: Flow rate of the zinc solution

Inlet ion concentration in the solution of Fe$^{2+}$, Fe$^{3+}$, H$^+$, respectively

Outlet ion concentration in the solution of Fe$^{2+}$, Fe$^{3+}$, H$^+$, respectively

$c_{O_2}$: Concentration of dissolved oxygen in the solution
Problem Formulation

- **Mechanism model of iron precipitation process**

  The mechanism model is established based on the mass balance and reaction dynamics of oxidation, hydrolysis and neutralization.

  \[
  \begin{aligned}
  \frac{dc_{Fe^{2+}}}{dt} &= \frac{F}{V} (c_{Fe^{2+},in} - c_{Fe^{2+}}) - k_1 c_{Fe^{2+}} c_{H^+} c_{O_2}^\gamma \\
  \frac{dc_{Fe^{3+}}}{dt} &= \frac{F}{V} (c_{Fe^{3+},in} - c_{Fe^{3+}}) + k_1 c_{Fe^{2+}} c_{H^+} c_{O_2}^\gamma - k_2 c_{Fe^{3+}} \\
  \frac{dc_{H^+}}{dt} &= \frac{F}{V} (c_{H^+,in} - c_{H^+}) - k_1 c_{Fe^{2+}} c_{H^+} c_{O_2}^\gamma + k_2 c_{Fe^{3+}} - \frac{3m_{ZnO}}{\rho R_s} k_3 c_{H^+} \\
  \frac{dc_{O_2}}{dt} &= k_{la} \left( \int \frac{\rho_{O_2} u_{O_2}}{M_{O_2} V} dt - 2c_{O_2} - \frac{1}{4} (c_{Fe^{2+},in} - c_{Fe^{2+}}) \right)
  \end{aligned}
  \]

  \(k_{la}\) is the output of a unmodeled dynamics.

  Data-based methods are used to build the model.

  \(\rho: \) Particle density of zinc oxide  

  \(R_s: \) Particle radius of zinc oxide  

  \(\rho_{O_2}: \) Density of oxygen  

  \(M_{O_2}: \) Molar mass of oxygen  

  \(u_{O_2}: \) Flow rate of oxygen  

  \(m_{ZnO}: \) Mass of zinc oxide  

  \(k_{la}: \) Mass transfer coefficient of oxygen  

  \(k_1, k_2, k_3, \alpha, \beta, \gamma: \) Parameters to be identified
Outline

- Introduction
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Main Results

- Hybrid modeling method for iron precipitation process

**Fig. 5 Hybrid model of the iron precipitation process**

$x$: States
$y$: Outputs
$\hat{y}$: Predictive outputs
$u$: Controls of mechanism model
$v$: Controls of data-based model
$f_1, f_2, g$: Functions
$C$: Output matrix
$
\xi$: Unmodeled dynamics
$\theta_1, \theta_2, \theta_3$: Parameters
hybrid model
Main Results

Hybrid model of iron precipitation process

- The hybrid model of iron precipitation process includes:
  - a mechanism model
    \[ \dot{x} = f_1(x, \theta_1, k_{la}) + g(x, \theta_2, k_{la})u \]
    \[ y = Cx, \]
  - and a data-based model of unmodeled dynamics \( k_{la} \)
    \[ k_{la} = f_2(x, u, v, \theta_3). \]

\[ x = [c_{Fe^{2+}}, c_{Fe^{3+}}, c_{H^+}, c_{O_2}]^T \]
\[ y = c_{Fe^{2+}} \]
\[ u = [u_{O_2}, m_{ZnO}]^T \]
\[ C = [1, 0, 0] \]

\( f_1 \) and \( g \): Can be obtained by some mathematical manipulation of the original mechanism model.

\( f_2 \): Need to be established based on data.
Main Results

Determine the model inputs of the mass transfer coefficient ($k_{la}$)

- $k_{la}$ is affected by following factors:
  1. the concentration of metal ions ($Fe^{2+}, Fe^{3+}, Cu^{2+}, Zn^{2+}$ and so on) in the solution
  2. the solids in the solution (goethite—$FeOOH$ and $ZnO$)
  3. solution flow rate and other factors

$$k_{la} = f_2\left(c_{Fe^{2+}}, c_{Fe^{3+}}, c_{Cu^{2+}}, c_{Zn^{2+}}, F, n_{FeOOH}, m_{ZnO}\right)$$

- $c_{Cu^{2+}}$: Concentration of $Cu^{2+}$
- $c_{Zn^{2+}}$: Concentration of $Zn^{2+}$
- $F$: Flow rate of the zinc solution
- $n_{FeOOH}$: Molar number of the goethite
- $m_{ZnO}$: Mass of zinc oxide
Main Results

- **Data-based modeling method for** \( k_{la} \)

  - \( k_{la} \) has strong nonlinearities with input variables. Different input variables have different correlations to \( k_{la} \).

  - Kernel principal component analysis (KPCA) and least squares support vector machine (LSSVM) are effective in dealing with strong process nonlinearities.

  - Locally weighted techniques can deal with correlations among different variables.

  - By incorporating the merits of KPCA, LSSVM and locally weighted techniques, a double locally weighted kernel principal component analysis-least squares support vector regression (DLWKPCA-LSSVR) is proposed to build the model of \( k_{la} \).
Main Results

- **Modeling method of DLWKPCA-LSSVR**
  
  **Step 1:** Determine input and output
  
  \[
  x = \left[ c_{Fe^{2+}}, c_{Fe^{3+}}, c_{Cu^{2+}}, c_{Zn^{2+}}, F, n_{FeOOH}, m_{ZnO} \right], \quad y = K_{La}
  \]

  Assume that the input variables of \( H \) historical samples used for modeling are \( \{x_h\}_{h=1}^H \), and the corresponding output samples are \( \{y_h\}_{h=1}^H \).

  **Step 2:** Calculate weights of historical samples

  The distance between each historical sample \( x_h \) and the query sample \( x_q \) (the real time sampling point) is calculated to obtain the weight of the historical sample.

  \[
  D_h = \sqrt{(x_h - x_q)^T (x_h - x_q)}, \quad h = 1, 2, \ldots, H
  \]

  \[
  w_h = \exp\left(-\frac{D_h^2}{\sigma^2}\right), \quad h = 1, 2, \ldots, H
  \]

  - \( D_h \): Distance between every historical sample and query sample
  - \( w_h \): Weight of the historical sample
  - \( \sigma \): Distance parameter
Main Results

Step 3: Calculate weights of input variables

The correlation coefficient:

\[ r = \frac{E(xy) - E(x)E(y)}{\sqrt{E(x^2) - E^2(x)} \sqrt{E(y^2) - E^2(y)}} \]

The weight of each dimension of the input variable:

\[ \lambda_s = |r_s| \sqrt{\sum_{k=1}^{L} |r_k|}, s = 1, 2, ..., L \]

The weighted input sample is:

\[ x_i^p = x_i \cdot \text{diag}(\lambda_1, \lambda_2, \ldots, \lambda_L) = [\lambda_1 x_{i1}, \lambda_2 x_{i2}, \ldots, \lambda_L x_{iL}]^T \]
Main Results

➢ Step 4: Extract nonlinearity by KPCA

The output-related nonlinearity is:

\[ g_q^{Pw,K} = K_q^{Pw} \alpha_d^{PW,K} \]

where

\[ K_q^{Pw} (q,i) = w_i K^P (q,i) \]

\[ K^P (i, j) = \varphi(x_i^P)^T \varphi(x_j^P) = e \frac{\|x_i^P - x_j^P\|^2}{2\delta_1^2} \]

- \( g_q^{Pw,K} \): Nonlinearity
- \( \alpha_d^{PW,K} \): First \( d \) columns of the eigenvector of \( K_q^{Pw} \)
- \( K^P \): Weighted kernel matrix
- \( \delta_1 \): A parameter
Main Results

- **Step 5:** Construct the LSSVR model between the output variable and the nonlinearity

\[ k_{la} = \sum_{i=1}^{N} \theta_i K(G_i^{Pw,K}, g) + b_N \]

where

\[ K(i, j) = e^{-\frac{\|x_i - x_j\|^2}{2\delta_2^2}} \]

- \( G_i^{Pw,K} \): Nonlinearity of training data
- \( g \): Nonlinearity of query sample
- \( K \): Kernel matrix
- \( \delta_2 \): Width parameter in kernel function
- \( N \): Number of modeling samples
- \( b_N \): Deviation that can be obtained by solving matrix equation
- \( \theta_i \): Lagrangian multiplier

By double locally weighting of samples and variables, we can extract the nonlinearity more related to the output.
Main Results

- Hybrid model of the process

\[
\begin{align*}
\frac{dc_{Fe^{2+}}}{dt} &= \frac{F}{V} (c_{Fe^{2+},in} - c_{Fe^{2+}}) - k_1 c_{Fe^{2+}}^\alpha c_{H^+}^\beta c_{O_2}^\gamma \\
\frac{dc_{Fe^{3+}}}{dt} &= \frac{F}{V} (c_{Fe^{3+},in} - c_{Fe^{3+}}) + k_1 c_{Fe^{2+}}^\alpha c_{H^+}^\beta c_{O_2}^\gamma - k_2 c_{Fe^{3+}} \\
\frac{dc_{H^+}}{dt} &= \frac{F}{V} (c_{H^+,in} - c_{H^+}) - k_1 c_{Fe^{2+}}^\alpha c_{H^+}^\beta c_{O_2}^\gamma - k_2 c_{Fe^{3+}} - \frac{3m_{ZnO}}{\rho R_s} k_3 c_{H^+} \\
\frac{dc_{O_2}}{dt} &= k_{la} \left( \int \frac{\rho_{O_2} u_{O_2}}{M_{O_2} V} dt - 2c_{O_2} - \frac{1}{4} (c_{Fe^{2+},in} - c_{Fe^{2+}}) \right)
\end{align*}
\]

The mechanism model

The data-based model

\[k_{la} = \sum_{i=1}^{N} \theta_i K(G_{i}^{Pw,K}, g) + b_N\]
Main Results

- Parameters identification

\[
\min J(\theta') = \frac{1}{N_{\text{train}}} \sum_{i=1}^{N_{\text{train}}} [(y' - \hat{y}')^2]
\]

\(\hat{y}'\) : Hybrid model predictive output

\(y'\) : Samples of the real output

\(\theta'\) : Parameter vector

\(N_{\text{train}}\) : Number of training samples

\[\theta' = [k_1, k_2, k_3, \alpha, \beta, \gamma, \sigma, \delta_1, \delta_2]\]

A global optimization algorithm – State Transition Algorithm is used to optimize the hybrid model parameters.
Main Results

Online updating strategy based on approximately linear dependence (ALD)

The ALD condition is used to determine whether to update the model when a new sample is available. The model updating condition is as follows.

\[
\begin{align*}
\delta_q &= \min \left\{ \sum_{i=1}^{N} a_i x_i - x_q \right\}^2 \\
\delta_q &\leq u, \quad \text{do not update the model} \\
\delta_q &> u, \quad \text{update the model}
\end{align*}
\]

- \( x_q \): New query sample
- \( x_i \): Training sample
- \( a_i \): Coefficient
- \( \delta_q \): ALD index
- \( u \): Given threshold

The parameters \( b_N \) and \( \theta_i \) in data-based model are updated when the ALD index reaches the given threshold.
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An Example

Taking 1# reactor of iron precipitation process as an example.

Model accuracy of three kinds of models are tested for comparison.

Model 1: The mechanism model in which $k_{la}$ is acquired by parameter identification

Model 2: A hybrid model in which $k_{la}$ is acquired by locally weighted kernel principal component regression (LWKPCR)

Model 3: The proposed hybrid model

Performance indices:

\[
\begin{align*}
RMSE &= \sqrt{\frac{\sum_{i=1}^{N_{test}} (y_i - \hat{y}_i)^2}{N_{test}}} \\
RMSE: \text{ Root mean squared error} \\
MAE &= \frac{\sum_{i=1}^{N_{test}} |y_i - \hat{y}_i|}{N_{test}} \\
MAE: \text{ Mean absolute error} \\
MRE &= \frac{1}{N_{test}} \sum_{i=1}^{N_{test}} \left| \frac{y_i - \hat{y}_i}{y_i} \right| \quad i = 1, 2, \ldots, N_{test} \\
MRE: \text{ Mean relative error}
\end{align*}
\]
An Example

Parameter setting

\[ \rho R_s : 0.012 \text{g/cm}^3 \]
\[ \rho O_2 : 1.429 \text{g/L} \]
\[ M_{O_2} : 32 \text{g/mol} \]
\[ V : 300 \text{m}^3 \]
\[ N : 10 \]
The principal components : 3

Parameter identification result for the hybrid model

Table 1 Parameter identification result

<table>
<thead>
<tr>
<th>Parameter</th>
<th>k_1</th>
<th>k_2</th>
<th>k_3</th>
<th>( \alpha )</th>
<th>( \beta )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Value</td>
<td>1.5963</td>
<td>0.0013</td>
<td>20.0868</td>
<td>1.3592</td>
<td>1.2807</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Parameter</th>
<th>( \gamma )</th>
<th>( \delta_1 )</th>
<th>( \delta_2 )</th>
<th>( \sigma )</th>
<th>( c )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Value</td>
<td>0.3764</td>
<td>4.4953</td>
<td>3.8011</td>
<td>1.9341</td>
<td>0.6491</td>
</tr>
</tbody>
</table>
An Example

- Model accuracy comparison of the three models

Fig. 6 The output of Model 1, Model 2, Model 3 and real system
An Example

➤ Model accuracy comparison results

Table 2 Comparison results on model accuracy of the three models

<table>
<thead>
<tr>
<th>Method</th>
<th>RMSE(g/L)</th>
<th>MAE(g/L)</th>
<th>MRE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Model 1</td>
<td>0.4747</td>
<td>0.3829</td>
<td>0.0458</td>
</tr>
<tr>
<td>Model 2</td>
<td>0.3816</td>
<td>0.3259</td>
<td>0.0406</td>
</tr>
<tr>
<td>Model 3</td>
<td>0.2315</td>
<td>0.1745</td>
<td>0.0219</td>
</tr>
</tbody>
</table>

The results in Table 2 show that the proposed hybrid model (Model 3) can better represent the dynamical characteristics of the iron precipitation process than the others.
An Example

The results of online updating strategy based on ALD

Fig. 7 Trend plot of computation time and RMSE with ALD threshold

The ALD threshold can be set to 0.185, which not only ensures the model accuracy, but also reduces the computation time.
Conclusion

- A hybrid model method by combining both a mechanism model and a data-based model of the mass transfer coefficient of oxygen is proposed for iron precipitation process.
  
  The mechanism model is established based on the mass balance and reaction dynamics.
  The unmodeled dynamics of the mass transfer coefficient of oxygen in mechanism model is built by DLWKPCA-LSSVR.

- Parameters in the hybrid model are identified simultaneously by using an optimization algorithm.

- An online updating strategy is proposed to reduce the computation time by setting a reasonable ALD threshold.
Conclusion and Future Work

Future Work

- Hybrid modeling method for industrial systems with the incomplete data.

- Adaptive parameter updating method for a hybrid model with various production conditions.
Thank you for your attention!