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**

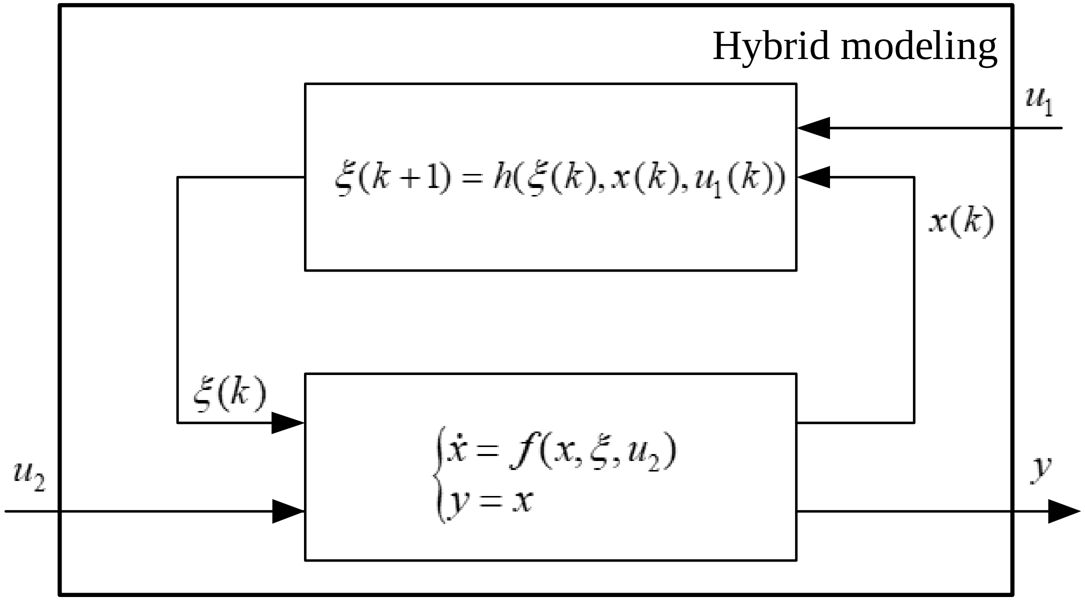
  Regression analysis between input and output variables

  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



x: States
y: Outputs
u_1,u_2: Controls
ξ: Unmodeled dynamics

Fig.1 Framework of a hybrid model

- 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.

Introduction

Modeling methods for iron precipitation by goethite

- A mechanism model of the iron precipitation process based on the reaction kinetics and mass balance

(F.Q. Xiong, W.H. Gui, C.H. Yang, et al, Journal of Central South University (Science and Technology), vol. 43, pp. 541-547, 2012.)

- An integrated model of the iron precipitation process by combining a mechanism model with an error compensation strategy

(Y.F. Xie, S.W. Xie, X.F. Chen, et al, Hydrometallurgy, vol. 151, pp. 62-72, 2015.)

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.

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Problem Formulation

The iron precipitation by goethite

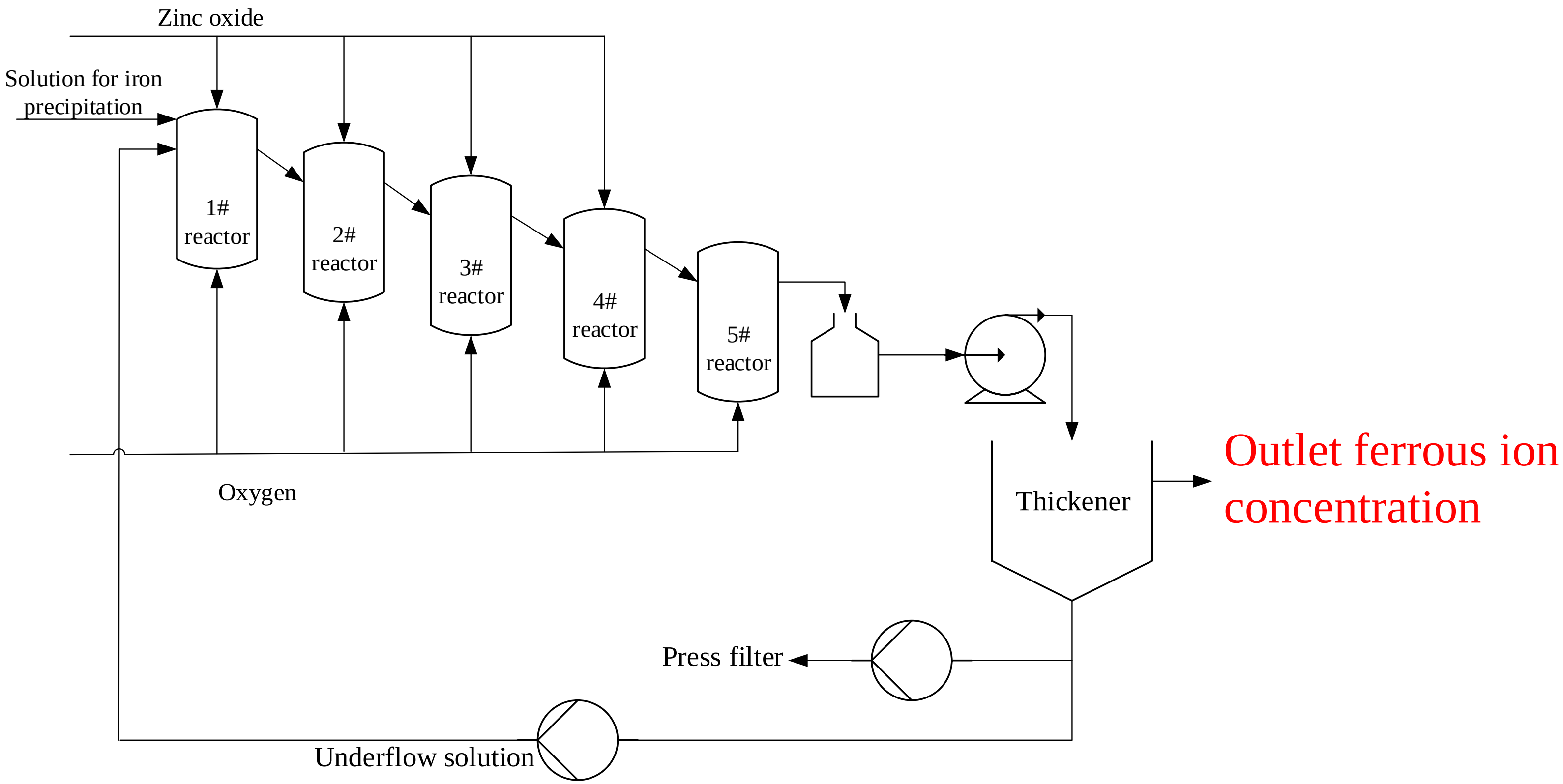

Fig.2 The process of iron precipitation by goethite

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

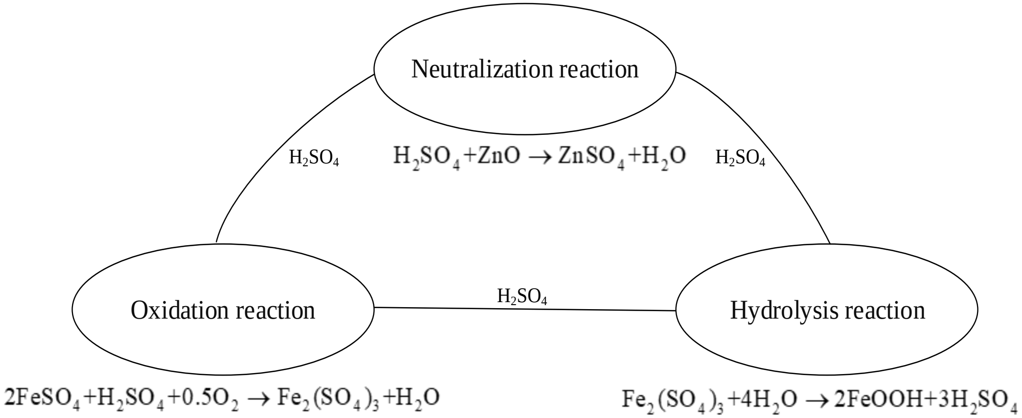

Fig.3 Three coupled reactions in iron precipitation

- 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

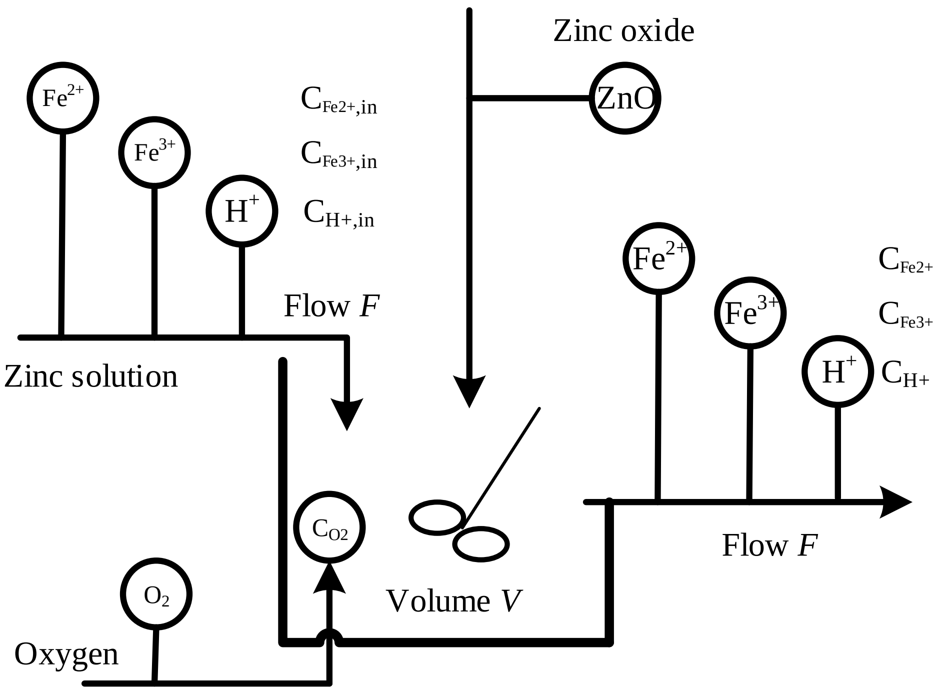

Fig.4 The reaction unit of the #1 reactor

V : Volume of the reactor

F: Flow rate of the zinc solution

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

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

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

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

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.

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

ρ: Particle density of zinc oxide

R_s : Particle radius of zinc oxide

ρ_{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

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

Data-based methods are used to build the model.

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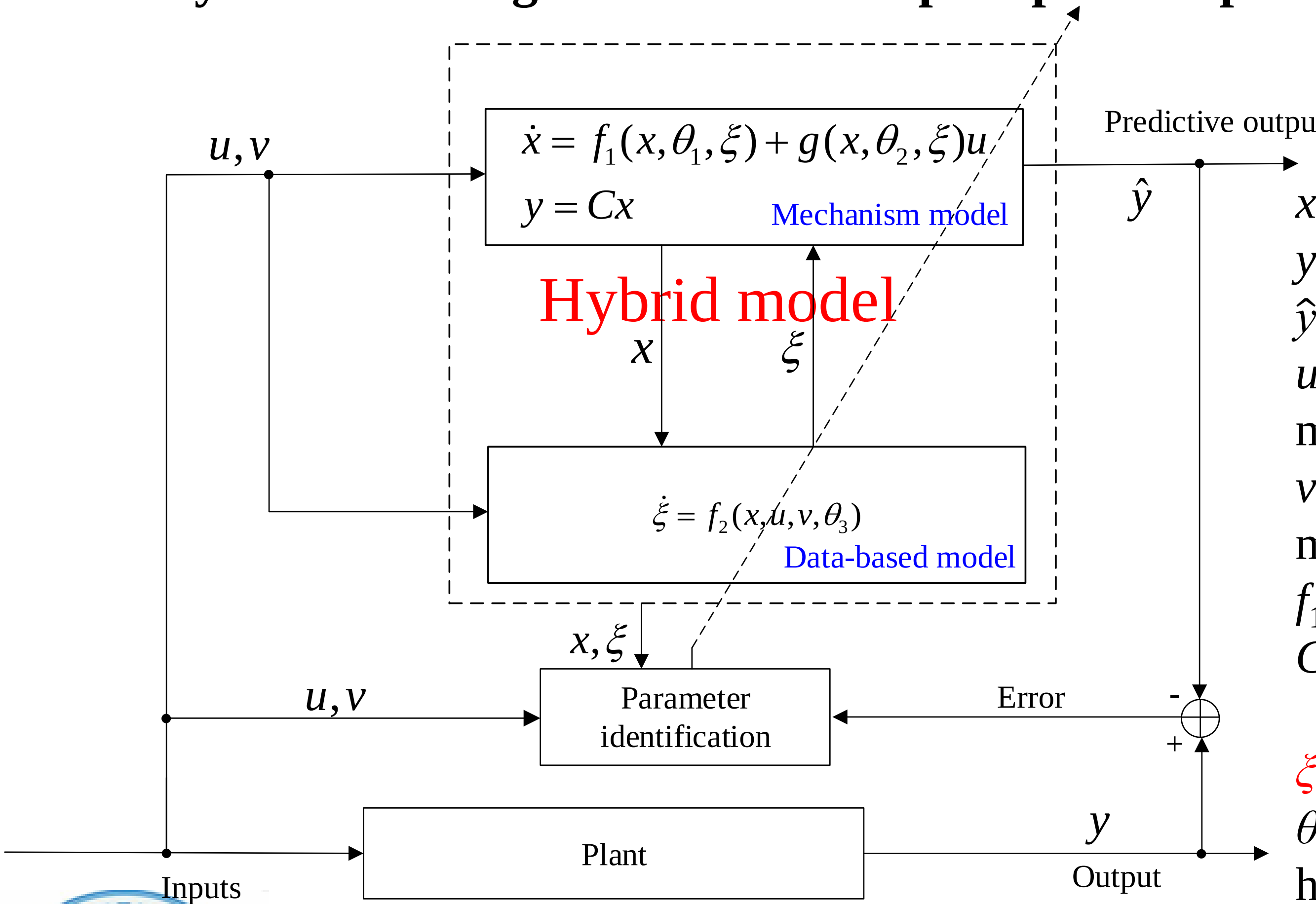
Main Results

Hybrid modeling method for 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

ξ: Unmodeled dynamics

$\theta_1,\theta_2,\theta_3$: Parameters hybrid model

Fig.5 Hybrid model of the iron precipitation process

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}

$$k_{la} = f_2(x,u,v,\theta_3).$$

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

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

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

$c_{Cu^{2+}}$: 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

$$\boldsymbol{x} = \left[c_{Fe^{2+}}, c_{Fe^{3+}}, c_{Cu^{2+}}, c_{Zn^{2+}}, F, n_{FeOOH}, m_{ZnO} \right], \quad \boldsymbol{y} = K_{La}$$

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 $\boldsymbol{x}_h$ and the query sample $\boldsymbol{x}_q$ (the real time sampling point) is calculated to obtain the weight of the historical sample.

$$D_h = \sqrt{(\boldsymbol{x}_h - \boldsymbol{x}_q)^T (\boldsymbol{x}_h - \boldsymbol{x}_q)}, h = 1, 2, \ldots, H$$

$$w_h = \exp(-D_h^2 / \sigma^2), h = 1, 2, \ldots, H$$

D_h: Distance between every historical sample and query sample

w_h: Weight of the historical sample

σ: 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:

$$\lambda_s = |r_s| \Big/ \sum_{k=1}^{L} |r_k|, s = 1, 2, ..., L$$

The weighted input sample is:

$$\boldsymbol{x}_i^p = \boldsymbol{x}_i \cdot diag(\lambda_1, \lambda_2, \ldots, \lambda_L) = [\lambda_1 x_{i1}, \lambda_2 x_{i2}, ..., \lambda_L x_{iL}]^{\mathrm{T}}$$

r : Pearson coefficient
E : Expectation
L: Dimension of input variable
λ_s: Weight of the input
x_{is}: s-th input of the i-th sample

Main Results

- Step 4： Extract nonlinearity by KPCA

The output-related nonlinearity is:

$$g_q^{Pw,K} = \boldsymbol{K}_q^{Pw} \boldsymbol{\alpha}_d^{PW,K}$$

where $\boldsymbol{K}_q^{Pw}(q,i) = w_i \boldsymbol{K}^P(q,i)$

$$\boldsymbol{K}^P(i,j) = \varphi(\boldsymbol{x}_i^p)^T \varphi(\boldsymbol{x}_j^p) = e^{-\frac{\left\| x_i^p - x_j^p \right\|^2}{2\delta_1^2}}$$

$g_q^{Pw,K}$: Nonlinearity

$\boldsymbol{\alpha}_d^{PW,K}$: First d columns of the eigenvector of $\boldsymbol{K}_q^{Pw}$

$\boldsymbol{K}^P$: Weighted kernel matrix

δ_1: A parameter

Main Results

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

$$k_{la} = \sum_{i=1}^{N} \theta_i \boldsymbol{K}(\boldsymbol{G}_i^{Pw,K}, \boldsymbol{g}) + b_N$$

where

$$\boldsymbol{K}(i, j) = \mathrm{e}^{-\frac{\left\|x_i - x_j\right\|^2}{2\delta_2^{\;2}}}$$

$\mathbf{G}_i^{Pw,K}$: Nonlinearity of training data

$\mathbf{g}$: Nonlinearity of query sample

$\boldsymbol{K}$: Kernel matrix

δ_2: Width parameter in kernel function

N: Number of modeling samples

b_N : Deviation that can be obtained by solving matrix equation

θ_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**

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

The mechanism model

$$k_{la} = \sum_{i=1}^{N} \theta_i \boldsymbol{K}(\boldsymbol{G}_i^{Pw,K}, \boldsymbol{g}) + b_N$$

The data-based model

Main Results

Parameters identification

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

$\hat{y}'$: Hybrid model predictive output

y' : Samples of the real output

θ' : Parameter vector

N_{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.

$$\begin{cases} \delta_q = \min \left\| \sum_{i=1}^{N} a_i x_i - x_q \right\|^2 \\ \delta_q \le u, \quad \text{do not update the model} \\ \delta_q > u, \quad \text{update the model} \end{cases}$$

x_q : New query sample
x_i : Training sample
a_i : Coefficient
δ_q : ALD index
u : Given threshold

The parameters b_N and θ_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:

$$\begin{cases} RMSE = \sqrt{\sum_{i=1}^{N_{test}} (y' - \hat{y}')^2 \Big/ N_{test}} \\ MAE = \sum_{i=1}^{N_{test}} \left| y' - \hat{y}' \right| \Big/ N_{test} \\ MRE = \dfrac{1}{N_{test}} \sum_{i=1}^{N_{test}} \left| \dfrac{y' - \hat{y}'}{y'} \right| \quad i = 1, 2, \ldots, N_{test} \end{cases}$$

RMSE: Root mean squared error
MAE: Mean absolute error
MRE: Mean relative error

An Example

- Parameter setting

ρR_s: 0.012g/cm^3
ρ_{O_2} : 1.429g/L
M_{O_2}: 32g/mol
V: 300m^3
N: 10
The principal components : 3

- Parameter identification result for the hybrid model

Table 1 Parameter identification result

Parameter	k_1	k_2	k_3	α	β
Value	1.5963	0.0013	20.0868	1.3592	1.2807
Parameter	γ	δ_1	δ_2	σ	c
Value	0.3764	4.4953	3.8011	1.9341	0.6491

An Example

- Model accuracy comparison of the three models

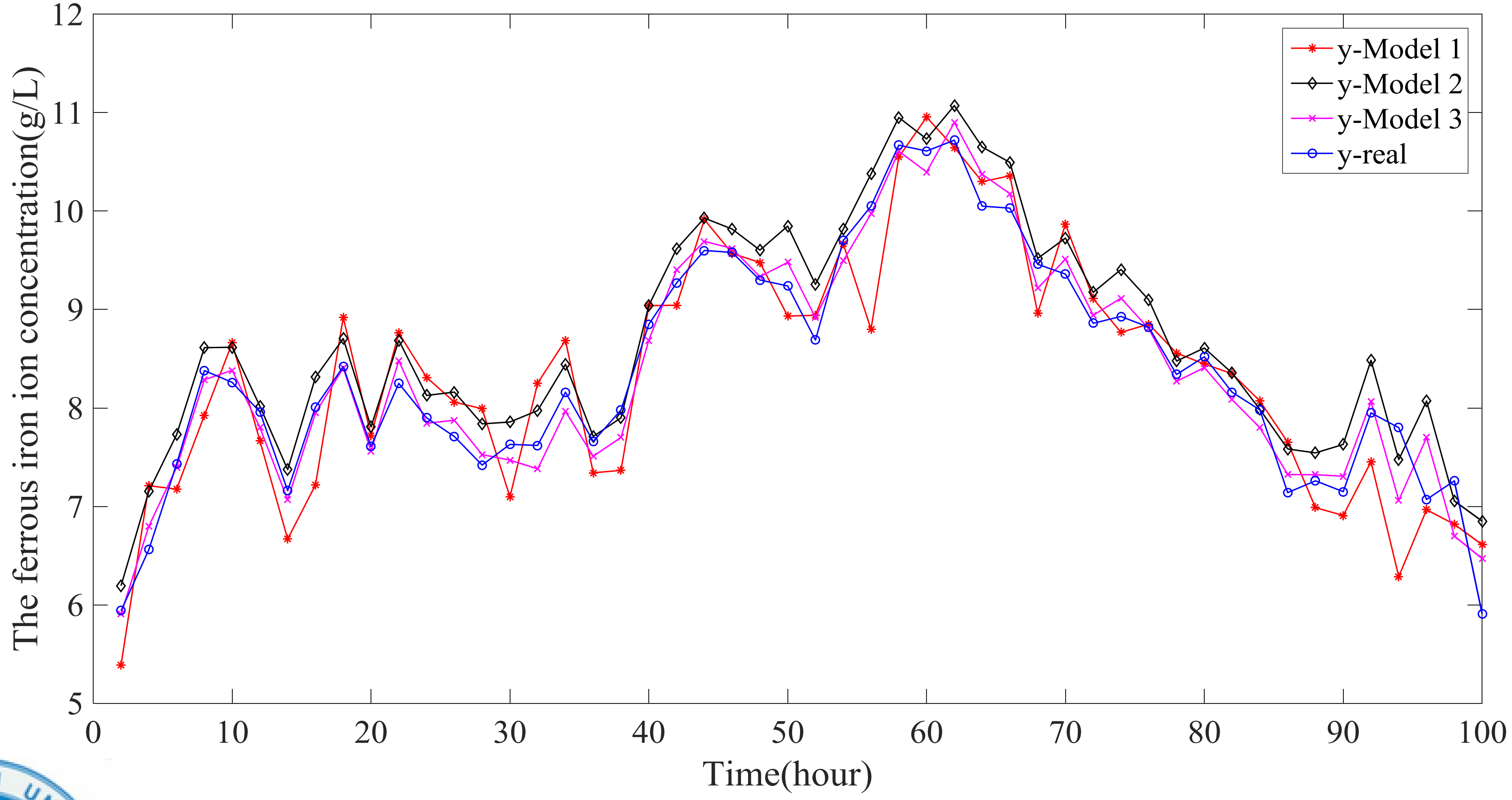

Fig.6 The output of Model 1, Model 2, Model 3 and real system

An Example

- Model accuracy comparison results

Table 2 Comparison results on model accuracy of the three models

Method	RMSE(g/L)	MAE(g/L)	MRE
Model 1	0.4747	0.3829	0.0458
Model 2	0.3816	0.3259	0.0406
Model 3	0.2315	0.1745	0.0219

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

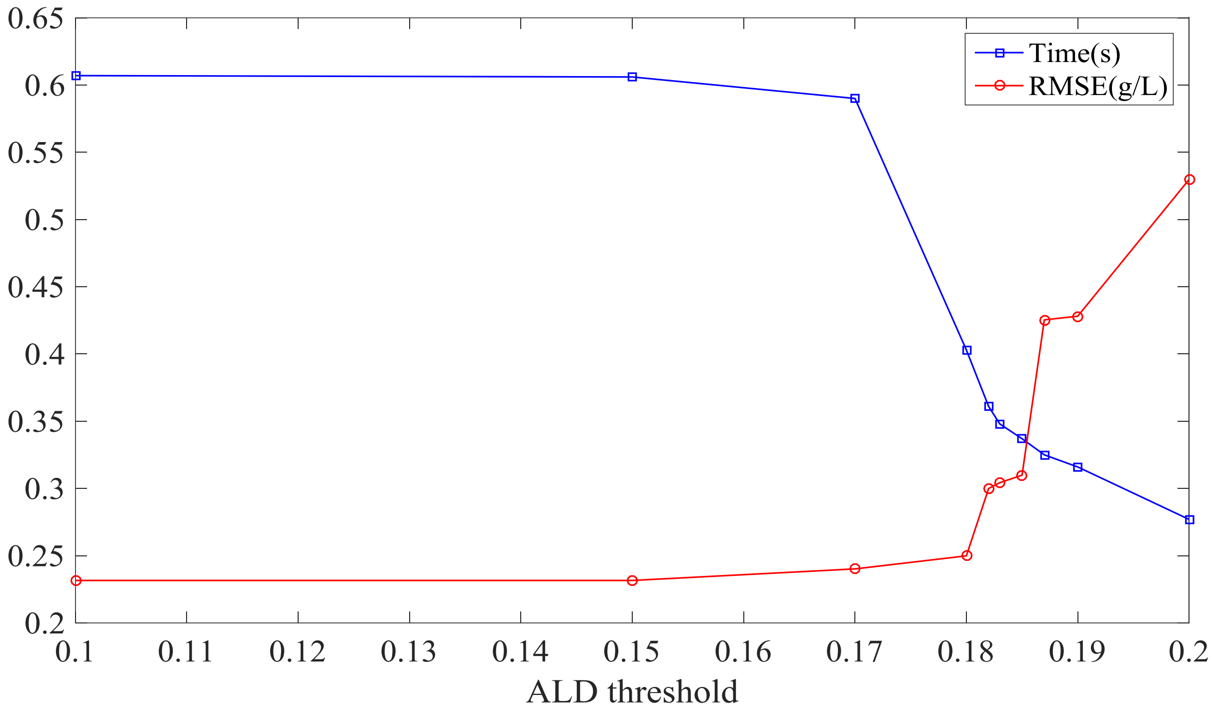

Fig.7 Trend plot of computation time and RMSE with ALD threshold

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

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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 !