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An integrated predictive model with an on-line updating strategy for iron precipitation in zinc hydrometallurgy

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ABSTRACT

Iron precipitation by goethite plays an important role in zinc hydrometallurgy. The ferrous ion concentration, which is a key index for assessing the iron removal rate and process control results, cannot be measured on-line. In this study, an integrated predictive model of the ferrous ion concentration is established by integrating the mechanism model and error compensation model, which is based on data identification. The mechanism model is proposed based on an analysis of the process reaction and considering the reaction unit as a continuous stirred tank reactor model. For unknown parameters in the mechanism model, a double-particle swarm optimization algorithm based on information exchange and dynamic adjustment of the feasible region is developed for optimal selection. To improve the adaptive capability of the integrated model, we propose a model-updating strategy and parameter calibration method based on a sensitivity analysis to accomplish on-line adaptive updating of the predictive model. The simulation results demonstrate that the proposed model can effectively track the variation tendency of the ferrous ion concentration and successfully improve the adaptability of the integrated model.

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1. Introduction

Zinc hydrometallurgy is the key technology in zinc production, which accounts for more than 80% of global zinc output (Balarini et al., 2008). Currently, the process of atmospheric direct leaching of zinc concentrate under oxygen-rich conditions is applied in most of the zinc hydrometallurgy industry. Because iron ions in the leaching solution interfere with zinc leaching and electrolysis, iron removal plays an important role in the zinc hydrometallurgy process. Iron precipitation in the form of goethite can reduce the total iron ion concentration of the leaching solution, which leads to better iron precipitate, greater crystallinity and easier filtering (Chang et al, 2010).

The process of removing iron by goethite is a complicated reaction in which the ferrous ion is oxidized to ferric iron by oxygen and the ferric iron is hydrolyzed to form the goethite polymer precipitate in a zinc sulfate solution (Claassen et al, 2002; Ismael and Carvalho, 2003). The reaction conditions have to be strictly controlled to obtain goethite precipitation, which means that the total iron ion concentration, the pH value and the temperature in the solution should be limited to a precise range. For example, 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, and the iron concentration at the reactor outlet might not satisfy the technical requirements. Furthermore, the ferrous ion concentration of every reactor outlet is an essential index for assessing the state of iron removal and controlling the operating variables. Because the iron precipitation procedure is long and complex, production operators manipulate it only according to their experience and the ferrous ion concentration of the reactor outlet. However, the ferrous ion concentration of the reactor outlet can only be assayed off-line with a long time delay, which increases the difficulty of real-time control of the iron removal process. Consequently, to predict the ferrous ion concentration on-line, establishing a dynamic model of iron precipitation is critical for stabilization and optimal guidance.

The procedure of iron precipitation by goethite usually includes several serial-connected reactors regarded as typical continuous-stirred tank reactors (CSTR). Currently, some modeling methods based on CSTR have been successfully applied to complicated industrial processes (Favache and Dochain, 2009; Gui et al., 2013; Li et al., 2012). For zinc hydrometallurgy, Haakana et al. (2007) investigated a novel reaction system for the direct leaching of zinc sulfide concentrates, and a detailed mathematical model for this system was developed. Markus et al. (2004) studied how to reduce dissolved ferric iron to ferrous ion with sphalerite concentrate, and the reaction kinetics were modeled. Sun et al. (2013) developed an integrated predictive model based on a kinetics model and a data-driven compensation model to predict cobalt ion concentration on-line in the cobalt removal process of zinc hydrometallurgy. As for the iron precipitation process, the chemical mechanism was investigated by

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many scholars (Formanek et al., 2013; Loan et al., 2006; Sergeeva et al., 2005). However, many of the methods are not of practical value in process modeling and predictive modeling. The reason is that the backflow solution in the iron precipitation process makes it an interconnected reaction system, which contains gas, liquid and solid phases in chemical reactions. The non-negligible interaction effect of chemical reactions increases the difficulty of modeling the process. Xiong et al. (2012) studied the reaction mechanism of the iron precipitation process, and established a dynamic model without considering the impact of copper ions. While in iron-removal chemical reactions, copper ions are a catalyzer for ferrous-ion oxidation, which improves the oxidizing speed. Thus, this dynamic model is insufficient to reflect the real process of iron precipitation. These previous works provide a good foundation for elaborating and building the predictive model of the ferrous ion concentration. A study on the modeling of the iron removal process by goethite can also be a reference for the paragoethite process or other zinc hydrometallurgy processes.

Considering the on-line predictive model of a complex industrial process the parameters of which are identified using industrial data, the variances existing in the industrial field and an input change will bring about a mismatch of model parameters and affect the prediction accuracy of the model. Concerning model-updating methods, Olivier and Craig (2013) studied the detection of parameter mismatch in a run-of-mine ore milling circuit model using a partial correlation analysis, and the model was updated by re-identification. He et al. (2013) proposed a model-updating method, in which the estimation of the parameter under the condition of complicated multi-physical fields was formulated as an optimization problem. Rui et al. (2013) developed a stochastic, model-updating method based on a surrogate model and implemented the updating of parameter mean values and variance. However, it is time-consuming to re-identify model parameters. The stochastic surrogate model discussed by Rui et al. (2013) cannot directly be applied to updating the complicated mechanism model of iron removal.

In this study, the mechanism of physicochemical reactions in the iron precipitation process by goethite is explored, and a mechanism model for iron removal is proposed to describe the changing law of the ion concentration in the reactors. To obtain a precise predictive model, a double particle swarm optimization algorithm based on information exchange (IEDPSO) is developed to identify the model parameters. Then, an error compensation model with a least squares support vector machine (LSSVM) trained by the historical error data is constructed to compensate for the shortcomings of the mechanism model. An integrated model that combines the mechanism model and the error compensation model is established. For the issue of parameter mismatch, a model parameter on-line calibration method based on a sensitivity analysis is proposed to improve the adaptive capability and reliability of the integrated model. It should be noted that this work differs from the author's previous works (Xie et al., 2014), where the present work focuses on modeling the process based on the reaction unit treated as a CSTR model and the on-line model updating method, which is very important to the on-line prediction model, while the previous work only built a prediction model of the ferrous ion concentration that simplified some important parameters. Thus, this paper can be considered to be an extension of our previous work.

The remainder of this paper is organized as follows. In Section 2, we analyze the process and give the model architecture based on the idea of integrated modeling. In Section 3, we present the mechanism model of iron precipitation at the beginning. Then, the model parameter identification algorithm based on a double particle swarm optimization of information exchange is introduced. An error compensation model is constructed based on the LSSVM algorithm. In Section 4, a model-parameter calibration method and a model-updating strategy are given and verified. The industrial application data are investigated to validate the efficiency of the proposed approach in Section 5. Section 6 gives some conclusions about the modeling method.

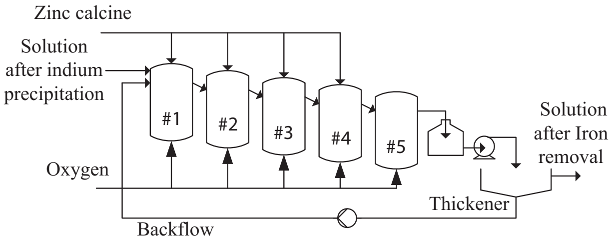

Fig. 1. The process of iron precipitation by goethite.

2. Process analysis and modeling strategy

2.1. Description of the iron precipitation process

In a certain zinc hydrometallurgy plant, the procedure for removing iron is to precipitate all of the iron ions in solution in the form of goethite after indium recovery. The technical process of iron precipitation is shown in Fig. 1, and some important procedure parameters or materials before iron removal are presented in Table 1. The system is composed of five reactors ranging from high to low according to the production flow. After indium precipitation, the zinc sulfate solution, including the main contents of Zn^{2+}, Fe^{2+}, Fe^{3+}, H^+, Cu^{2+} and other impurity ions, flows into the #1 reactor via a chute. The iron-removed solution from the #5 reactor flows into a thickener for solid–liquid separation. Part of the thickener underflow returns to #1 rector as a crystal seed. In these five reactors, ferrous ions are oxidized to ferric ions by atmospheric oxygen, and ferric ions are hydrolyzed to form goethite. To balance the pH, a neutralization agent with the main components of zinc calcine is added into the first four reactors.

The ferrous ion concentration of every reactor outlet is an important index for measuring the state of iron removal and as a reference for controlling the addition of oxygen and zinc calcine. Meanwhile, the ferrous ion concentration of the reactor outlet should be controlled within a certain range to obtain satisfactory goethite precipitation. In particular, the mass concentration of ferrous ion in the iron-removed solution should be less than 1 g/L to achieve the process requirements. However, the ferrous ion concentration is tested every 2 h such that the real-time information is not available to control the addition of oxygen and zinc calcine.

In the iron removal process, three main chemical reactions occur in reactors, including the oxidation reaction of ferrous ions, hydrolysis of ferric ions and a neutralization reaction. The chemical reactions in the five reactors are the same, and their reaction dynamic models are similar. Therefore, we take the #1 reactor as a case to study the reaction mechanism of the three chemical reactions. The differential unit shown in Fig. 2 is defined as the reaction unit for analyzing the chemical reactions in this process. With the material continuously being added into the reactor, the three reactions occur in a reaction unit simultaneously with the change in ion concentration. Due to radial agitation in the stirrer, the ion concentration is assumed to be uniform in the reaction unit at the same position. Therefore, every unit is a typical CSTR model. We analyze the reaction mechanism of three reactions in

Table 1
The process parameters or materials.

Materials	Range	Sample frequency
Inlet solution (m^3/h)	80–160	On-line measured
Backflow solution (m^3/h)	30–50	On-line measured
Fe^{2+} (g/L)	9–18	Off-line assayed every 2 h
Fe^{3+} (g/L)	1–3	Off-line assayed every 2 h
H^+ (pH)	2–3	On-line measured
Cu^{2+} (g/L)	1–2	Off-line assayed every 2 h
Oxygen flow (m^3/h)	300–500	On-line measured
Zinc calcine (t/h)	2–4	On-line measured

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