PARAMETER OPTIMAL IDENTIFICATION AND DYNAMIC BEHAVIOR ANALYSIS OF NONLINEAR MODEL FOR THE SOLUTION PURIFICATION PROCESS OF ZINC HYDROMETALLURGY

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Abstract. Impurity removal is a momentous part of zinc hydrometallurgy process, and the quality of products and the stability of the whole process are affected directly by its control effect. The application of dynamic model is of great significance to the prediction of key indexes and the optimization of process control. In this paper, considering the complex coupling relationship of stage II purification process, a hybrid modeling method of mechanism modeling and parameter identification modeling was proposed on the basis of not changing the actual production process of lead-zinc smeltery. Firstly, the overall nonlinear dynamic mechanism model was established, and then the deviation between the theoretical value and the actual detected outlet ion concentration was taken as the objective function to establish the parameter identification optimization model. Since the built model is nonlinear, it may pose implementation problems. On the premise of deriving the gradient vector and Hessian matrix of the objective function with respect to the parameter vector, an optimization algorithm based on the steepest descent method and Newton method is proposed. Finally, using the historical production data of a lead-zinc smeltery in China, the model parameters were accurately inverted. An intensive simulation validation and analysis of the dynamic characteristics about the whole model shows the accuracy and the potential of the model, also in the perspective of practical implementation, which provides the basis for the optimal control of system output and the guidance for the optimal control of zinc powder addition.

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1. Introduction. Zinc is a kind of light gray transition metal, which is often used as the raw material of precision castings, and it has an indelible position in the field of battery manufacturing in modern industry. Zinc hydrometallurgy has been developed rapidly due to its advantages of high metal recovery, low air pollution, low energy consumption, good working conditions, easy realization of mechanization and automation [9]. In the leaching process of zinc hydrometallurgy, the impurities in zinc sulfate leaching solution have great harm to the zinc electrolysis process, which will reduce the electrolytic current efficiency, increase the power consumption, affect the quality of cathode zinc and corrode the cathode. Therefore, the main impurity ions such as cadmium, cobalt, nickel and copper in neutral leaching solution must be removed to the allowable content range of electrolysis process through solution purification, so as to ensure the normal operation of electrolysis process and produce high-grade zinc sheet [21].

The best opportunity to control the process may be lost because the operators cannot get the change of ion concentration in time in the purification process of zinc hydrometallurgy. In addition, zinc powder, as the main raw material for impurity removal, is a quite significant control condition. Based on experience, excessive zinc powder is usually used to acquire the qualified solution by the operators. Although the purification quality of zinc sulfate solution is ensured by this method, the production cost and energy consumption are greatly increased [2]. Therefore, on the premise of ensuring the continuous production of qualified electrolytic solution, reducing the consumption of zinc powder, decreasing the production cost and improving the economic benefit has become a problem to be solved in zinc hydrometallurgy production, and the realization of the prediction of the outlet ion concentration of stage II purification process is the basis to solve this problem.

The impurity removal process is a multi-reactor cascade process including various physical and chemical reactions, which has the characteristics of strict process conditions and extremely complex internal dynamics. For the control of impurity removal and similar complex industrial process, scholars at home and abroad have carried out numerous research work. B. Sun et al. established a data-driven model based on support vector machine to solve the problem of insufficient description ability and adaptive ability of the kinetic model, and combined it with the kinetic model. The results showed that the proposed integrated model can accurately predict the cobalt ion outlet concentration in time [6]. In 2015, the idea of gradient optimization of solution purification process was put forward, which transformed the economic optimization of solution purification process into the problem of finding the optimal descending gradient of impurity ion concentration along the reactors [7]. The following year, using the online measurement characteristics of the oxidation-reduction potential, an online estimation method of impurity ion concentration was proposed, which simultaneously utilized domain knowledge, offline daily production data and online data [8]. The impurity ion coupling relationship in removal process was not considered in the above literature. S. W. Xie et al. proposed a two-step parameter identification method. The first production data was used to identify the model parameter vector, while the second production data was used to identify the weight vector. The results showed that the method has high prediction accuracy [18]. A spectrophotometric method combining continuous wavelet transform with zero-crossing technology was developed to determine cobalt and copper in zinc hydrometallurgy by Ultraviolet-visible spectroscopy without any separation steps by F. B. Zhou et al. [19]. Aiming at the difficulty of online detection of inlet ion
concentration at stage II, a soft sensing method of ion concentration parameters based on wavelet analysis and support vector machine was proposed by H. Q. Zhu. The soft sensing model of ion concentration was directly established by using the historical data of manual detection. The verification results showed that the method can satisfy the actual production process requirements [20]. In [19] and [20], the accuracy of process data is very high. According to the complex characteristics of cobalt removal process by arsenic salt, a collaborative optimization method of operation parameters based on fuzzy operation mode was proposed by T. B. Wu, which solved the problem of difficult optimization of operation parameters [14].

X. Y. Wang used statistical method to analyze the distribution characteristics of uncertain parameters, introduced the idea of chance constraint, and modeled the optimization problem of copper removal process under uncertain conditions as a chance constrained optimization problem. Finally, the sequential quadratic programming was used to solve the nonlinear programming problem [15]. The method is relatively difficult to apply to practice. In addition, the problem of impurity removal in zinc hydrometallurgy is also studied in literature [16, 17, 11, 4].

Stage II purification is the most critical part in the whole purification process. Among the current achievements, either the method requires too high accuracy of process historical data; or the method is highly dependent on expert experience, which is difficult to be applied to practice; or the coupling relationships of impurity ions in removal process are not been considered. In view of the problems existing in the research on the optimization control of the impurity removal process in the above literature, combined with the current actual process of a lead-zinc smeltery in China, the mechanism modeling for stage II purification process of zinc hydrometallurgy is carried out on the basis of not changing the actual production process, and then the deviation between the theoretical value and the actual detected outlet ion concentration is taken as the objective function to establish the parameter identification optimization model. Because the mechanism model cannot be divided into linear form, the gradient vector and Hessian matrix of the objective function with respect to the parameter vector are derived, and the optimization algorithm based on the steepest descent method and Newton method is given. The unknown parameters in the mechanism model are inversed by using the parameter identification model, and then the nonlinear dynamic behavior characteristics of the purification process are analyzed. The dynamic influence of the model variables on the output during the operation of the system is clarified, and the corresponding simulation diagrams are obtained to verify the effectiveness of the model and realize the prediction of the outlet ion concentration for the stage II purification process, which provides the basis for the optimal control of the system output, and is of great significance for the in-depth study in the actual solution purification process of zinc hydrometallurgy.

The remainder of this paper is organized as follows. Section 2 describes the stage II purification process and establishes the mechanism model. Section 3 establishes the parameter identification optimization model and gives the solving algorithm. Combined with a lead-zinc smeltery in China, the analyses of model dynamic behavior and model validity are shown in Section 4. The conclusions are given in Section 5.
2. Plant description and modeling.

2.1. Description of stage II purification process. The target of purification process is zinc sulfate leaching solution from zinc concentrate [1]. The flow chart of roasting, leaching and purification process of a lead-zinc smeltery is shown in Fig. 1. Firstly, zinc concentrate is selected from zinc-containing minerals, and then to burn zinc concentrate into zinc calcine in roasting furnace. The dilute sulfuric acid solution or electrolytic spent solution from electrolysis process is used as solvent to dissolve zinc from zinc calcine.

The whole purification process is divided into three stages. The neutral supernatant from the leaching process enters stage I purification process. At the solution temperature of 50 ∼ 60 ℃, zinc powder is added in the first reaction tank to remove cadmium ion and copper ion. After the stage I purification process, the concentration of impurity ions in the solution can generally reach a relatively stable state. After pressure filtration and heating, the outlet solution of stage I enters the stage II purification process. At temperature of 87 ∼ 90 ℃, zinc powder and antimony salt are added in the first reaction tank of stage II purification to remove cobalt ion and continue to remove cadmium ion. After pressure filtration, the outlet solution of stage II enters the purification process of stage III, and a small amount of zinc powder is added to remove the redissolved cadmium ion.

Among the many impurity ions, cobalt ion not only affects the efficiency of electrolysis, but may also cause electrolytic burnout when exceeding the standard seriously. Therefore, the key parameter of cobalt ion concentration in the purification process must be strictly monitored. Meanwhile, the content of cadmium ion is much higher than that of other components, which is also a momentous technical index for the purification of cadmium ion concentration. Consequently, the cobalt ion and cadmium ion in the solution are mainly analyzed and predicted in this paper, which corresponds to the stage II purification process. The process is composed of 5 reactors in cascade. The reaction solution flows downstream from the former reactor to the next reactor. The sufficient chemical reaction is carried out under the stirring of 1#~5# continuous reactors to achieve the aim of removing cobalt and cadmium in turn. The average time of the purification reaction in this process is about 2 hours, and the reactions that occur mainly include:

\[
\begin{align*}
\text{Sb}^{3+} + \text{Co}^{2+} + 2.5\text{Zn} &= \text{CoSb} \downarrow + 2.5\text{Zn}^{2+} \\
\text{Sb}^{3+} + \text{Cd}^{2+} + 2.5\text{Zn} &= \text{CdSb} \downarrow + 2.5\text{Zn}^{2+}
\end{align*}
\]

The ion concentration of the solution during stage II purification is relatively low. The amount of zinc powder can be adjusted by the operators according to the concentration of cobalt ion and cadmium ion detected at the inlet and outlet of stage II. Because there is a certain time delay in the determination of ion concentration, if the unqualified ion concentration information is acquired by the operators, it means that a batch of unqualified solution have entered the next process, which is extremely unfavorable to the production.

The purification effect of stage II is affected by many factors, including temperature, reaction time, mixing degree of powder particles and solution, concentration of impurity ions such as cobalt and cadmium in the solution after stage I, addition amount of zinc powder and antimony salt, composition and particles size of zinc powder, etc., which affect the rate of displacement reaction in varying degrees, and then affect the reaction process of removing cobalt ion and cadmium ion [12].
2.2. Modeling of stage II impurity removal process. The consumption of zinc powder in the stage II purification process is related to the impurity content at the outlet of stage I, especially the concentration of cobalt ion and cadmium ion, as well as the chemical composition and particles size of zinc powder and technical conditions such as the temperature of solution. This paper is based on the actual production process of a lead-zinc smeltery, so the process conditions are not changed, that is, the chemical composition and particles size of zinc powder remain unchanged. Blowing zinc powder is used in the stage II purification process, in which the zinc element accounts for 95% and the particle size is 200 mesh. The two 8-hour temperature profiles in the same period of time on different dates of stage II solution purification in a lead-zinc smeltery are show in Fig. 2. It can be seen from the figure that in the stage II purification process, the temperature fluctuates between 87°C and 89°C. Meanwhile, during the purification process, no solution outside the system is injected into the solution of the reaction tank as a coolant. Consequently, in the process of modeling, it can be considered that the temperature remains unchanged and its impact on the system is ignored. The concentration of cobalt ion and cadmium ion at the outlet of stage I and the amount of zinc powder added are the main factors affecting the process of zinc powder replacement and impurity removal.

The continuous stirred tank reactor (CSTR) is a kind of reactor widely used in metallurgical and chemical production process, which is a highly nonlinear chemical reaction dynamic system. The CSTR model is a typical mechanism model of nonlinear reaction kinetics, which is used to describe the critical processes of hydrometallurgical reaction and metal ion purification in nonferrous metallurgical process [3]. It is assumed that the material flows into the reactor at a steady flow rate, and the material and the material remaining in the reactor reach complete mixing instantaneously in CSTR mechanism modeling.
Fig. 2. Temperature profiles of stage II solution purification in a lead-zinc smeltery

In the actual zinc smeltery, the ion concentration at the inlet of 1# reaction tank and the outlet of 5# reaction tank is mainly detected, and the amount of zinc powder is often adjusted according to these data. The solution and solid particles in the series-combined reactor model can be completely mixed. Therefore, the original chemical reaction process can still be simulated by taking the five series reactors as a whole CSTR when the mathematical model is established, as shown in Fig. 3.

The purification of zinc sulfate solution is a long time delay process. The solution starts from the inlet of 1# reaction tank of stage II purification and needs to stay in the reaction tank for an average of 2 hours before it flows to the outlet of the 5# reaction tank. Consequently, after considering the 5 reaction tanks as a whole CSTR, the ion concentration of the outlet solution actually reflects the change of the ion concentration 2 hours ago.

Using zinc powder to replace different impurity ions in zinc sulfate solution, the whole replacement process is coupled and interfered with each other. The degree of this mutual influence is positively related to its concentration in the solution. The greater the concentration of interfering ions is, the greater the impact on other ions is [13].

To explore the relationship between the internal variables in the stage II purification process of zinc hydrometallurgy, the main manifestation is the use of differential equations to describe the input-output relationship and the interaction between the internal variables. According to the above principle analysis of impurity removal process for zinc hydrometallurgy, on the basis of material balance, the nonlinear dynamic reaction mechanism model by zinc powder replacement to remove cobalt ion and cadmium ion in stage II purification process can be established, as shown in formula (1):

\[
\begin{align*}
\frac{dx_a(t)}{dt} &= \frac{Q}{V} (x_{a0} - x_a(t-2)) - \frac{k_a}{V} \cdot p \cdot u_a(t)x_a(t-2) + \frac{\alpha}{V} x_b(t-2) \\
\frac{dx_b(t)}{dt} &= \frac{Q}{V} (x_{b0} - x_b(t-2)) - \frac{k_b}{V} \cdot p \cdot u_b(t)x_b(t-2) + \frac{\beta}{V} x_a(t-2)
\end{align*}
\]
Inlet solution
Zinc powder &
antimony salt
Outlet solution

Figure 3. The equivalent CSTR model

Where, \( x_a(t) \) and \( x_b(t) \) represent the concentration of cobalt ion and cadmium ion at the outlet of reaction tank respectively; \( x_{a0} \) and \( x_{b0} \) are the concentration of cobalt ion and cadmium ion at the inlet of reaction tank respectively; \( x_a(t - 2) \) and \( x_b(t - 2) \) mean the concentration of cobalt ion and cadmium ion at the outlet before 2 hours respectively; \( u_a(t) \) and \( u_b(t) \) represent the weight of zinc powder used to remove cobalt ion and cadmium ion respectively; \( Q \) and \( V \) are the flow rate and volume of solution respectively; \( p \) means the coefficient between reaction surface area and weight of zinc powder (assuming that zinc powder particles are tiny spheres); \( k_a \) and \( k_b \) represent chemical reaction coefficients respectively; \( \alpha \) and \( \beta \) are ion coupling coefficients respectively, where \( \alpha \) indicates the influence of cadmium ion on cobalt ion, and \( \beta \) indicates the influence of cobalt ion on cadmium ion.

The input variables involved in the dynamic mechanism model include the inlet solution flow rate and zinc powder addition. According to the analysis of production data, although the inlet solution flow rate will fluctuate in a certain range, the average flow rate is basically stable. Consequently, the amount of zinc powder is selected as the control variable to optimize the concentration of impurity ions at the outlet.

3. Parameter identification optimization model.

3.1. Parameter identification modeling. In Section 2, the overall nonlinear dynamic mechanism model of stage II purification process is established, and the basic structure of the model is determined. However, there are still four unknown parameters \( \chi = (k_a, k_b, \alpha, \beta)^T \) to be identified in the model. Accordingly, the parameters in the model will be optimized in this section to obtain a complete hybrid dynamic model.

The basic idea of parameter identification is to minimize the deviation between theoretical calculation and actual measurement by modifying the nominal value of parameters [5]. Here, the theoretical value is expressed as the function of the state vector \( S \) and the parameter vector \( \chi = (k_a, k_b, \alpha, \beta)^T \), and the deviation function as shown in formula (2) can be defined to transform the identification problem into the minimum problem of the objective function.
\[ e(S) = \sum_{j=1}^{N} [x_a(S_j, \chi) - \bar{x}_{aj}]^2 + [x_b(S_j, \chi) - \bar{x}_{bj}]^2 \]  

Where, \( S_j \) is the \( S \) vector measured at the \( j \)th sampling point; \( \bar{x}_{aj} \) and \( \bar{x}_{bj} \) are the concentration of cobalt ion and cadmium ion at the outlet measured at the \( j \)th sampling point respectively; \( N \) is the total number of sampling points in the process of parameter identification.

The optimization model of parameter identification can be expressed as:

\[ \chi^* = \arg \min_{\chi} e(\chi) \]  

Where \( \chi^* \) is the optimal parameter vector.

3.2. Solving algorithm of parameter identification optimization model.

Since the theoretical value \((x_a, x_b)\) is not a linear form, it cannot be solved by the least square method. Consider using the following iterative method to solve the optimization model in formula (3):

\[ \chi^{(k)} = \chi^{(k-1)} + \Delta \chi \]  

Where, the superscript \( k \) represents the number of iterations; \( \Delta \chi \) is the parameter increment vector of each iteration.

Generally, the increment \( \Delta \chi \) is determined by Newton iteration method:

\[ \Delta \chi = -H^{-1}G \]  

Where \( G \) and \( H \) are the gradient vector and Hessian matrix of the error function \( e \) respectively. The steepest descent method shown in formula (6) is used to solve \( \Delta \chi \) is when the Hessian matrix \( H \) is singular.

\[ \Delta \chi = -\alpha G \]  

Where \( \alpha \) is the learning rate (\( \alpha > 0 \)). The convergence condition of the above iterative process can be expressed as:

\[ ||\Delta \chi|| \leq \varepsilon_1 \]  

Where \( \varepsilon_1 \) is the preset error limit. The formula (4) \(~ (7)\) is synthesized, and the specific calculation flow of the algorithm for solving the above optimization model is shown in Fig. 4.

The Hessian matrix \( H \) and gradient vector \( G \) in the above algorithm are respectively:

\[ H = \frac{\partial^2 e}{\partial \chi^2} = 2 \sum_{j=1}^{N} \left\{ \frac{\partial^2 x_a}{\partial \chi^2} [x_{aj} - \bar{x}_{aj}] + \frac{\partial^2 x_b}{\partial \chi^2} [x_{bj} - \bar{x}_{bj}] + \left[ \frac{\partial x_a}{\partial \chi} \right] ^T \left[ \frac{\partial x_a}{\partial \chi} \right] + \left[ \frac{\partial x_b}{\partial \chi} \right] ^T \left[ \frac{\partial x_b}{\partial \chi} \right] ^T \right\} \]  

\[ G = \frac{\partial e}{\partial \chi} = 2 \sum_{j=1}^{N} \left\{ \frac{\partial x_a}{\partial \chi} (x_{aj} - \bar{x}_{aj}) + \frac{\partial x_b}{\partial \chi} (x_{bj} - \bar{x}_{bj}) \right\} \]  

Where, \( x_{aj} = x_a(S_j, \chi), x_{bj} = x_b(S_j, \chi) \).
4. Results and discussion. The part to be identified in this paper is the stage II purification process of zinc hydrometallurgy in a lead-zinc smeltery in China. The reaction process of this stage is composed of 5 reaction tanks, and some actual photos are shown in Fig.5. The relevant parameters of the reaction process are shown in Table 1.
Table 1. Values of relevant parameters in the reaction process

| Parameter                                      | Symbol | Value |
|------------------------------------------------|--------|-------|
| Solution flow rate / (m³/h)                   | Q      | 160   |
| Volume of single reaction tank / m³           | V_p   | 108   |
| Volume utilization of reaction tank / %       |        | 80    |
| Area coefficient / (m²/kg)                    | p      | 174   |

Using the historical data of the lead-zinc smeltery in October 2020, 200 samples are selected to calculate the optimal parameters according to the parameter identification algorithm given in Section 3. The characteristics of the selected sample data are shown in Table 2.

Table 2. Characteristics of sample data

| Data type                                      | Symbol | Average value | Maximum value | Minimum value |
|------------------------------------------------|--------|---------------|---------------|---------------|
| Inlet cobalt ion concentration / (mg/L)        | x_{a0} | 35.251        | 51.190        | 13.382        |
| Inlet cadmium ion concentration / (mg/L)       | x_{b0} | 298.278       | 433.148       | 113.229       |
| Outlet cobalt ion concentration / (mg/L)       | x_{a}  | 0.439         | 0.945         | 0.257         |
| Outlet cadmium ion concentration / (mg/L)      | x_{b}  | 15.772        | 58.943        | 1.449         |

In the process of solving, the learning rate $\alpha$ is determined by the trial and error process. The initial value is set to 1, that is, the adjustment of parameter vector $\chi$ is determined according to 100% gradient vector, and then the value of $\alpha$ is gradually reduced until the three consecutive calculations can converge to the same result. At this time, it is considered that the parameter identification algorithm has achieved stable convergence. The corresponding deviation function profile and parameter increment profile are shown in Fig. 6. After 76 iterations, the algorithm converges stably. At this time, the deviation function value is 0.0997, and $\|\Delta \chi\| = 8.92 \times 10^{-5}$. In this paper, the value of $\alpha$ corresponding to the stable convergence of the algorithm is 0.03, and the value of $\varepsilon_1$ is set to $10^{-4}$. After solving the identification model, the optimal model parameters obtained are $\chi^* = (7.352 \times 10^{-4}, 5.963 \times 10^{-4}, 8.32, 860)^T$.

4.1. Analysis of model dynamic behavior. According to the above description of the mathematical model in stage II purification process of zinc hydrometallurgy, the given differential equations and the identified parameters are used in this section to build the stage II purification process model in Matlab / Simulink. The output performance will be affected by the changes of input and structural parameters of the system. The relationship between these variables is studied by simulation. The stability of the model is tested by adding step input to verify the correctness of the model and the influence of process parameters on the system. Some conclusions of great significance to the optimization control of stage II purification process are obtained. The model is shown in Fig. 7. The inlet ion concentration values are shown in Table 3.
Figure 6. Deviation function profile and parameter increment profile in solving process.(a)Deviation function profile;(b)Parameter increment profile

Table 3. Inlet ion concentration values

| Inlet ion          | Symbol | Value |
|--------------------|--------|-------|
| Cobalt ion / (g/L) | $x_{a0}$ | 0.035 |
| Cadmium ion / (g/L)| $x_{b0}$ | 0.298 |

According to the actual production data, the ratio of zinc powder consumed in the process of removing cobalt ion and cadmium ion in the stage II purification process is about 1:5, so in the simulation model, set $u_a : u_b = 1 : 5$. Fig. 8 shows the response profiles of cobalt ion and cadmium ion concentration at the
outlet of stage II purification process when the amount of zinc powder (control variable) changes stepwise under the condition of inlet solution flow of 160m³/h. It can be seen from Fig. 8(a) that when the amount of zinc powder is given, the outlet cobalt ion concentration increases continuously for about 10s, and the final cobalt ion concentration value tends to be stable. Given different amount of zinc powder, the profiles of cobalt ion concentration at the outlet have a similar trend, but the final stable values are different. It can be seen that the stable value of cobalt ion concentration at the outlet is not positively correlated with the amount of zinc powder. It is not that the larger the amount of zinc powder, the smaller the concentration of cobalt ion at the outlet. The same conclusion can be drawn from Fig. 8 (b).

It can also be seen from Fig. 8 (a) and Fig. 8 (b) that with the change of $u_a$, the concentration of cobalt ion at the outlet is constantly changing. Due to the mutual coupling between ions, the concentration of cadmium ion at the outlet is affected, and this change is almost simultaneous. In a certain range, the larger the $u_a$ is, the smaller the cobalt ion concentration at the outlet is, and the smaller effect on the cadmium ion concentration at the outlet is. Similarly, with the constant change of $u_b$, the concentration of cadmium ion at the outlet changes, which affects the concentration of cobalt ion at the outlet, and the larger the $u_b$ is, the smaller effect of cadmium ion concentration on the concentration of cobalt ion is. It shows that different impurity ions are coupled and interfered with each other in the whole replacement process. The higher the concentration of interfering ions, the greater the influence on other ions.

In order to further illustrate the stability of the built model and the coupling effect between ions, the input signals $u_b$ (Fig. 9) and $u_a$ (Fig. 10) are designed. Assuming that one input signal is constant, the influence of the other input on the output is observed. Fig. 9 shows the variation profiles of cobalt ion concentration (a) and cadmium ion concentration (b) at the outlet when $u_b$ is constant and $u_a$ changes; Fig. 10 shows the variation profiles of cobalt ion concentration (a) and cadmium ion concentration (b) at the outlet when is constant and changes; Fig. 11 shows the variation profiles of cobalt ion concentration (a) and cadmium ion concentration (b) at the outlet when is constant and changes;
Fig. 8 shows the influence of the inlet flow rate of stage II purification process on the concentration of cobalt ion and cadmium ion in the outlet solution under the same operating conditions and input conditions. It can be seen from Fig. 12(a) that when the inlet flow rate of stage II is 0.8Q, 1.2Q respectively, the rising trend of cobalt ion concentration at the outlet of stage II is different, and finally the stable output state of the system is reached. The greater the inlet flow rate is, the smaller the outlet ion concentration tends to equilibrium. The outlet ion concentration has a downward trend at 6s when the inlet flow rate is 1.2Q. The reason is that...
Figure 9. Variation profiles of outlet ion concentration when $u_\theta$ is constant and $u_a$ is variable. (a) Cobalt ion concentration at the outlet; (b) Cadmium ion concentration at the outlet.
Figure 10. Variation profiles of outlet ion concentration when $u_a$ is constant and $u_b$ is variable. (a) Cobalt ion concentration at the outlet; (b) Cadmium ion concentration at the outlet.
under the condition of constant control variable of zinc powder, with the increase of inlet flow rate, the time to reach the reaction volume is shortened, the reaction area between zinc powder and impurity ions is increased, the decreasing time of outlet ion concentration is advanced, and the steady-state value is also decreased. However, when the inlet flow rate is 0.8 $Q$, the ion concentration at the outlet does not decrease after reaching the maximum value, and the maximum value is equal to the steady-state value, which results in model mismatch. This is because the reaction coefficient and coupling coefficient are identified when the inlet flow is $Q$, and the model parameters are not suitable for the case when the inlet flow is 0.8$Q$. The change trend of the cadmium ion concentration at the outlet of stage II in Fig. 12(b) has the same situation as the change trend of the cobalt ion concentration.

According to the above analysis of the principle, structure and related performance simulation for solution purification process of zinc hydrometallurgy, it can be found that solution purification process of zinc hydrometallurgy involves many complex processes, which is a nonlinear system with multiple inputs, multiple outputs and easily disturbed.

4.2. Analysis of model validity. In Section 4.1, the dynamic behavior of the model is analyzed, which shows that different impurity ions are coupled and interfered with each other in the whole replacement process when the amount of zinc powder (control variable) changes. Meanwhile, it shows that the hybrid dynamic reaction model is a stable process.

Using the collected data, the real production process is simulated, and 80 samples are selected to test the identification results. The test results are shown in Fig. 13. The accuracy of the model can be verified by comparing the calculated data with the measured data. Accordingly, the average relative deviation between the
Figure 12. The influence of inlet flow $Q$ on impurity ions concentration. (a) Cobalt ion concentration at the outlet; (b) Cadmium ion concentration at the outlet.

calculated results of the model and the measured data is defined as the simulation error of the model in this paper [10], and the expression is shown in formula (10):

$$
\varepsilon_{2a}\% = 100 \times \frac{\sum_{j=1}^{M} \left( \frac{x_{a,j} - \bar{x}_{a,j}}{\bar{x}_{a,j}} \right)}{M}
$$

$$
\varepsilon_{2b}\% = 100 \times \frac{\sum_{j=1}^{M} \left( \frac{x_{b,j} - \bar{x}_{b,j}}{\bar{x}_{b,j}} \right)}{M}
$$

Where, $\varepsilon_{2a}\%$ is the simulation error of cobalt ion at the outlet; $\varepsilon_{2b}\%$ is the simulation error of cadmium ion at the outlet; $M$ is the number of samples included in the measured data. The smaller $\varepsilon_2\%$ is, the closer the prediction results are to the measured data, the higher the accuracy and prediction accuracy of the model are,
and the better the prediction effect is. Table 4 shows the simulation error comparison results between the nonlinear dynamic model in this paper and the model in reference [8].

| Error                               | Model in this paper | Original model [8] | Reformulated model [8] |
|-------------------------------------|---------------------|--------------------|------------------------|
| Maximum error (Cobalt ion)/%       | 31.12               | 49.10              | 32.27                  |
| Maximum error (Cadmium ion)/%      | 24.40               | \                 | \                      |
| Average error (Cobalt ion)/%       | 11.05               | 13.81              | 11.90                  |
| Average error (Cadmium ion)/%      | 10.85               | \                 | \                      |
It can be seen that the average prediction relative error of the nonlinear dynamic model in this paper is less than 11%, which verifies the high accuracy of the model and the identified parameters in the stage II purification of zinc hydrometallurgy. Compared with the original model and the reformulated model in reference [8], there is a smaller relative error with this method.

5. Conclusions. An accurate dynamic model is one of the prerequisites to obtain a good control effect. Consequently, before controlling the purification process of zinc hydrometallurgy, modeling and parameter identification are often required to reduce the error of theoretical model in the controller. The dynamic mechanism model and parameter identification optimization model of stage II purification process established in this paper constitute the overall nonlinear dynamic model. Aiming at the problem that the model cannot be divided into linear form, the optimization algorithm based on steepest descent method and Newton method is derived and given. Finally, through the actual data of a lead-zinc smeltery in China, the parameters of the model are inversed. Simulation examples show that based on the model and parameters in this paper not only have excellent stability, which shows that different impurity ions are coupled and interfered with each other in the whole replacement process, but also can accurately fit the historical data of stage II purification process, reveal the reaction mechanism of stage II purification process, effectively predict the ion concentration at the outlet of stage II purification process, and provide the basis and guidance for the subsequent optimization of the controller design.

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