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Distributed parameter modeling and optimal control of the oxidation rate in the iron removal process

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ABSTRACT

Generally, the iron removal process is modelled as a lumped parameter system that does not provide information about the distribution of reactants in the steady state. In this paper, we investigate the distributed parameter model and control for the iron removal process. By analyzing the process properties, we study the mass balance over a differential volume element, and the spatiotemporal distributions of the Fe^{2+}, Fe^{3+} and H^{+} concentrations are derived by partial differential equations. An optimization problem is constructed to estimate the unknown parameters. Then, an optimal control problem for the oxidation rate of the ferrous ions in the steady state is proposed to achieve process requirements that have the lowest cost of oxygen and zinc oxide and obtain high goethite quality. To eliminate the impact from inevitable disturbances, an expert-based correction mechanism is constructed to compensate for the optimal control when the outlet ferrous ion concentrations are out of the desired range. Finally, the simulation results demonstrate the good performance of distributed parameter model. Industrial experiments demonstrate the satisfactory control performance of the optimal control strategy. Regarding manual operation and PI control, the control strategy increased the qualified ratio of the #4 reactor outlet Fe^{2+} concentrations by 8.4% and 3.4%, respectively. Additionally, on average 17760 m^3 of oxygen and 109.68 t of zinc oxide per month were saved compared to manual operation. The mass percent of iron in the goethite increased from 34.31% (manual operation) and 35.12% (PI control) to 35.83%.

1. Introduction

The technology of iron removal as goethite has been widely applied in zinc hydrometallurgy plants [1–4]. The procedure removes iron ions by adding oxygen to oxidize the ferrous ions to ferric ions, and the latter is then hydrolyzed to form goethite under an acid environment. Zinc oxide should be added to the solution to maintain an appropriate acid environment. The iron removal process is a complicated industrial process that involves intricate chemical reactions. The main process requirements of the iron removal process are removing the iron ions to the desired range at the lowest cost for oxygen and zinc oxide and to obtain the goethite with as high a content of iron as possible.

To achieve the process requirements, efficient control of the oxygen and zinc oxide is essential. In addition, controlling the oxidation rate of ferrous ions is important because it is a main factor affecting the quality of the goethite precipitate. In practice, some important data, such as the ferrous ion concentration and ferric ion concentration, can only be measured by off-line assay, so we cannot obtain real-time feedback information to guide the control of oxygen and zinc oxide. The external disturbances, intricate process mechanism and limited information regarding the oxidation rate of ferrous ions also cause difficulties in controlling the iron removal process. Consequently, establishing a process model to predict the dynamic behavior of the ferrous ion concentration and designing an efficient control method for the iron removal process are important.

Over the past several years, the modeling methods applied to the industrial processes have generally focused on the continuously stirred tank reactor (CSTR) [5–7]. In particular, in the zinc hydrometallurgy leaching and purification process, the reactor is a typical continuously stirred tank reactor that all of the chemical species in the reactor are completely mixed and homogeneously dispersed [8]. In practice, many industrial processes belong to distributed parameter systems (DPSs) with significant spatio-temporal dynamics. The consideration that the process is a distributed parameter system offers more options for improving the observation capacity of the state variables [9]. Therefore, many modelling approaches for DPSs are documented in the literature [10–12]. A spatiotemporal least square support vector machine (LS-SVM) modelling method for complex nonlinear DPS

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was proposed that integrates the space kernel function and the time Lagrange multiplier [13]. This method was successfully applied in the curing thermal process. However, negligence in the reaction process and mass balance in the metallurgical process will cause the loss of some process properties. Owing to the limited controllers and computing power, model reduction is necessary for practical implementation [14], which means that the infinite-dimensional controllers in the DPS need to be approximated by a finite-dimensional controller. The time-space separation technique was proposed to reduce the modeling complexity [15,16]. In this technique, the spatio-temporal variables are separated into a set of spatial basis functions and temporal models, and through the time-space synthesis, the spatio-temporal system will be recovered. Its modelling performance depends on the selection of the spatial basis functions and system identification methods, which make the model not work well in a wide range of operating conditions. Owing to the nonuniform ore source of the zinc hydrometallurgy plant in China, numerous operating conditions are included in the iron removal process. Based on a previous study [17], mass balance is a good method to describe its distributed parameter model (DPM). However, motivated by the above literatures, it is crucial to reduce the model dimensions when establishing the DPM for the iron removal process and design a control strategy for practical implementation.

In the iron removal process, the oxidation rate of the ferrous ions is an important indicator to assess the process operational performance. To operate the process with high performance and guarantee the quality of the goethite precipitate, the oxidation rate of the ferrous ions should be restricted to a reasonable range [4]. Designing a control strategy to achieve the process requirements and control the oxidation rate is necessary. For a complex DPS, its nonlinearities and the limited availability of sensors cause many control difficulties. Many researchers have developed control methods for DPSs, such as the fuzzy control [18–21], output tracking control [14] and model predictive control (MPC) methods [22–25]. The fuzzy logic control method has been widely used in industrial processes because a satisfactory controller can be developed empirically without complex mathematics. However, because the fuzzy logic control method does not require the system model, MPC may be a more suitable method for industrial process with DPM. For a different DPS, the system properties and process requirements are different. Therefore, the objective function and constraints are different in their MPC scheme, such as in [22] and [24]. Consequently, the mentioned MPC cannot be directly used in the iron removal process. Inspired by the MPC scheme, an optimal controller for the iron removal process is developed to achieve the process requirements and control the oxidation rate of the ferrous ions.

To address the above challenges, we establish a distributed parameter model and design optimal control strategy for the iron removal process in zinc hydrometallurgy. Based on reasonable assumptions, the DPM is derived according to the mass balance over a differential volume element. For the unknown parameters in the model, the parameter estimation problem is transformed into an optimization problem, which is solved by a particle swarm optimization (PSO) algorithm. To improve the quality of the goethite precipitate, we construct an optimal control strategy to achieve the process requirements and control the oxidation rate based on the established DPM in the steady state. A correction mechanism is proposed based on expert knowledge and experience to eliminate the impact from inevitable disturbances and changes in production conditions. Finally, experiments are conducted to collect production data and identify the model parameters. The performance of the DPM is verified by the simulation results, and the optimal control strategy is demonstrated by industrial experiments and compared with manual operation and the PI control.

This paper is organized as follows. Section 2 describes and analyzes the iron removal process in zinc hydrometallurgy. The DPM and parameter estimations are presented in Section 3. The optimal control strategy is developed in Section 4. In Section 5, the simulations and industrial experiments are shown. In Section 6 conclusions are drawn.

2. Process description and analysis for iron removal by goethite

2.1. Description of iron removal process

Zinc hydrometallurgy is realized from zinc concentrate under oxygen-rich conditions using atmospheric direct leaching technology. The iron removal process as a goethite precipitate is an essential procedure in the atmospheric leaching process. This paper focuses on a certain iron precipitation procedure shown in Fig. 1.

This process includes four continuously stirred reactors and a thickener for solid-liquid separation. The inlet solution, which is a zinc sulfate solution, contains mainly Zn^{2+}, Fe^{2+}, Fe^{3+}, H^{+}, Cu^{2+}, and $SO_4{}^{2-}$. Approximately 90% of the iron ions are ferrous ions. The atmospheric oxygen is fed to the bottom of the reactors, and part of the oxygen is dissolved in the solution. The ferrous ions in the reactors are oxidized to ferric ions by the dissolved oxygen, and the ferric ions are precipitated as goethite. Zinc oxide (ZnO) powders are added to the solution from the top of the reactors. In the zinc sulfate solution, the hydrogen ions react with ZnO particles to balance the pH. Throughout the process, the solution pH should be maintained at approximately 3.0. The solution after iron removal is transferred to a thickener for separating the liquid and solids, and the goethite precipitate is obtained from the thickener underflow. Part of the thickener underflow is circulated to the #1 reactor for seeding purposes. The active volume of the reactor is approximately 300 m^3. In the iron removal process, the temperature of the solution should be maintained at approximately 353 K.

The three most important chemical reactions that occur in the reactors are described as follows. These reactions involve the gas, liquid and solid phases and are physically and chemically intricate. Therefore, the procedure is a heterogeneous system.

$$4Fe^{2+}+O_2+4H^{+} \rightarrow 4Fe^{3+} + 2H_2O \tag{1}$$

$$Fe^{3+}+2H_2O \rightarrow FeOOH \downarrow +3H^{+} \tag{2}$$

$$2H^{+} + ZnO \rightarrow Zn^{2+} + H_2O \tag{3}$$

Under the acidic environment and the catalytic action of Cu^{2+}, the Fe^{2+} is oxidized to Fe^{3+} which is hydrolyzed to a goethite precipitate. It is worth mentioning that the mass concentration of Fe^{3+} should not exceed 2 g/L to obtain a satisfactory goethite product rather than ferric hydroxide ($Fe(OH)_3$) [4]. The goethite precipitation process is achieved by slow oxidation of the ferrous ions. Therefore, the concentration of ferrous ions in the reaction process is a crucial feedback indicator to control the process. We can obtain the dynamic behavior of the ferrous ions from the DPM whether in a dynamic or steady state. Consequently, the DPM is essential for the control of the iron removal process.

2.2. Analysis of procedure

The content of the inlet solution is shown in Table 1. The mass concentration of Fe^{2+} is much larger than that of Fe^{3+}. If the reaction rate of the ferrous ions is too fast, the concentration of Fe^{3+} may exceed 2 g/L such that ferric hydroxide ($Fe(OH)_3$) will produce. On the contrary, if it is too slow, it is difficult to remove the ferrous ions to the required range. Hence, the reaction rate of Fe^{2+} is a key parameter in the process. A process requirements of the iron