Control Engineering Practice 58 (2017) 54–65

Contents lists available at ScienceDirect

Control Engineering Practice

journal homepage: www.elsevier.com/locate/conengprac

Dynamic modeling and optimal control of goethite process based on the rate-controlling step

Yong-fang Xie, Shi-wen Xie*, Yong-gang Li, Chun-hua Yang, Wei-hua Gui

School of Information Science and Engineering, Central South University, ChangSha City, 410083, China

ARTICLE INFO

Keywords:
Dynamic model
Rate-controlling step
Pre-setting of descent gradient
Optimal control
Goethite process

ABSTRACT

The iron removal process is an important technology in zinc hydrometallurgy. Due to its complicated reaction mechanism, it is difficult to highly control the performance of process solely relying on manual experience. Therefore, this paper focuses on the dynamic modeling for the goethite process and its optimal control method. In different reactors, different operating conditions will influence the rate-controlling step of process reactions, as well as the kinetic model. We determine the rate-controlling step of reactions in each reactor according to the reaction conditions, and subsequently the dynamic model has been developed based on the rate-controlling step. Then an optimal control method for the goethite process has been proposed, which satisfies the technical requirements with minimal process consumption. The proposed optimal control includes pre-setting of descent gradient of outlet ferrous ion concentration and optimal control of oxygen and zinc oxide. The simulation results demonstrate that the proposed dynamic model exhibits greater performance comparing with the process model without considering rate-controlling step, and the proposed control strategy has a higher satisfactory than the nonlinear model predictive control. Then, the proposed optimal control is validated by the industrial experiment, which the average oxygen and zinc oxide consumptions decreased by 568 m^3/day (6.22%) and 3.09 t/day (5.16%) and the qualified rate of outlet ferrous ion concentrations increased by 5.5%, compared with the manual control.

1. Introduction

In the zinc hydrometallurgy process, iron ions must be removed from the leaching solution in order to enhance the efficiency of zinc electrolysis and to improve the quality of the zinc product. Consequently, efficiently removing the iron ions is an essential step in the hydrometallurgical extraction of zinc. Currently, the dissolved iron can be precipitated in the form of jarosite, hematite and goethite, which are widely utilized in many smelting factories (Formanek, Jandova, & Capek, 2013; Loan, Newman, Cooper, Farrow, & Parkinson, 2006). The iron removal process by goethite, namely goethite process, is commercially significant, because it forms larger crystals and is consequently easier to filter. Goethite precipitate contains high concentrations of iron, which can be a mineral material in the smelting of steel after further treatment (Torfs & Vliegen, 1996).

In the goethite process, three main reactions are involved in the formation of goethite precipitate, including oxidation reaction, hydrolysis reaction and neutralization reaction. And they can be described by the following stoichiometric equations (Xie et al., 2015).

$$\text{Oxidation reaction: } 2Fe^{2+} + 0.5O_2 + 2H^{+} \rightarrow 2Fe^{3+} + H_2O \tag{1}$$

$$\text{Hydrolysis reaction: } Fe^{3+} + 2H_2O \rightarrow FeOOH + 3H^{+} \tag{2}$$

$$\text{Neutralization reaction: } 2H^{+} + ZnO \rightarrow Zn^{2+} + H_2O \tag{3}$$

It is observed that the goethite process contains three complicated chemical reactions and these reactions are interacted. For instance, the hydrogen ion is involved in three reactions. Consequently, it is difficult to describe the reaction process utilizing simple mathematical models. However, in order to accurately control the process and to ensure the procedure requirements, it is crucial for a dynamic model to be developed. If the model is accurately developed, some process indices that could be used as real-time feedback in the controlling of the procedure. Therefore, an analysis of the mechanism for iron precipitation reactions and the establishment of a dynamic model are required for the stabilization of the process, and the effective and efficient operation.

Many modeling methods in the field of chemical engineering have been studied and discussed in the literature, such as blending process in alumina metallurgical industry (Yang, Gui, Kong, & Wang, 2009), cobalt removal process (Dreher, Nelson, Demopoulos, & Filippou, 2001) and copper cementation (Demirkıran, Ekmekyapar, Künkül, &

* Corresponding author.
E-mail address: mathking@csu.edu.cn (S.-w. Xie).

http://dx.doi.org/10.1016/j.conengprac.2016.10.001
Received 3 December 2015; Received in revised form 6 September 2016; Accepted 7 October 2016

Baysar, 2007), etc. As to the iron removal process in zinc hydrometallurgy, the majority of the theoretical works have been previously discussed (Chang, Zhai, Li, & Fu, 2010; Claassen, Meyer, Rennie, & Sandenbergh, 2002; Ismael & Carvalho, 2003). Ismael and Carvalho (2003) suggested that the goethite process can be successfully implemented in the electrolytic industry, and it can be accomplished by the reduction of all of the ferric ions to a ferrous state or by the addition of a concentrated ferric solution. The chemical reactions and the reaction conditions of goethite precipitation were also given. Claassen et al. (2002) introduced the methods of iron removal from the zinc-rich solution and the nature of the goethite. The chemical removal of iron by goethite was given and some experiments were conducted in order to obtain goethite. Chang et al. (2010) reported the iron removal by goethite precipitation from the leach liquor, and some influences on the process were also investigated. All of previous studies provide the inspiration and the theoretical fundamentals for the dynamic modeling of the goethite process. This previous chemical researches play an important role in the analysis of process mechanism. However, these chemical theories did not determine the reaction rates under different operating conditions, which would affect the modeling of the diverse production states.

In general, a chemical reaction consists of multi-step reaction. According to the theory of reaction rate-controlling step, the reaction rate of the overall reaction is determined by the lowest rate of the sub-step reaction (Nazemi, Rashchi, & Mostoufi, 2011). It is crucial to find the lowest rate of the sub-step reaction, and calculate its reaction rate. Currently, in the modeling of the multi-reactor process in zinc hydrometallurgy, the existing researches have usually employed one model for the whole procedure or considered that the model established for one reactor is applicable to other reactors (Sun, Gui, Wang, & Yang, 2014; Zhang, Yang, Zhu, Li, & Gui, 2013). Nevertheless, the different operating conditions in different reactors will have influence on the reaction rate, which may be completely different, as well as on the dynamic model. Therefore, the analysis of reaction rate at different operational conditions is a key issue in the dynamic modeling for goethite process.

In this paper, our aim is to design an optimal control law such that the technical requirements are met at the cost of minimal consumption. In order to achieve the procedure requirements with least consumption, many approaches have been developed and applied to process control. Model predictive control (MPC) (De Souza, Odloak, & Zanin, 2010), fuzzy control (Li, Shi, Wu, & Zhang, 2014; Marsili-Libelli & Colzi, 1998), expert control (Chen, Li, & Fei, 2008; Zhang, Yang, Zhu, Li, & Gui, 2016), intelligent optimal control (Chai, Ding, & Wu, 2011), robust adaptive control (Sun, Gui, Wang, Yang, & He, 2015), etc are widely researched in the industrial process. In practice, for a particular metallurgy process, the above control methods are not commonly used. Additionally, it is difficult to directly use the existing control methods for the particular process with specific performance required and different constraints (Chai et al., 2011). Therefore, developing a specific control method for the goethite process is necessary.

In our study, the concept of reaction rate-controlling step is adopted in calculating the reaction rate. Firstly, a further analysis of the relationship between the oxygen concentration and the ferrous ion concentration in each reactor is made. It is contribute to calculate the reaction rate in the different operational condition, combining with the chemical theory of rate-controlling step. The dynamic model for each reactor in goethite process is established based on the rate-controlling step. The unknown parameters in the dynamic model are estimated by a modified particle swarm optimization (PSO) algorithm. Then, based on the process model, an optimal control method is developed to achieve the procedure requirements with minimal consumption. The control method is composed of pre-setting of descent gradient of outlet ferrous ion concentration and optimal control of oxygen and zinc oxide. The pre-setting part helps for constraining the oxidation rate of ferrous ion according to production conditions to guarantee the technical requirements, and gives the distinct targets for the optimal control. Finally, the established model and proposed control are verified by simulation and industrial application, respectively.

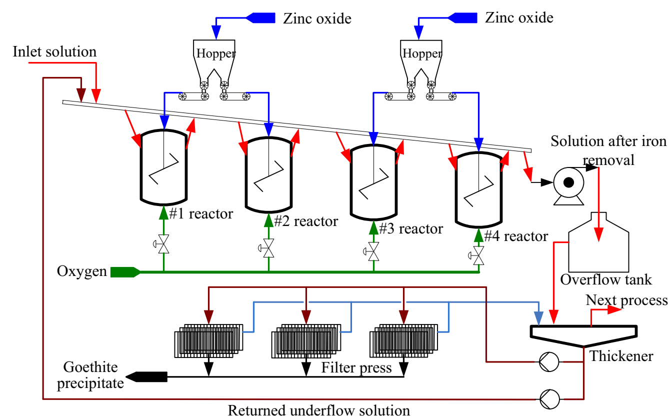

Fig. 1. Schematic of the goethite process.

The rest of this paper is organized as follows. Section 2 describes the goethite process. In Section 3, the rate-controlling step of each reaction in different reactors is analyzed and determined. The dynamic model is established and the unknown parameters are identified in Section 4. Section 5 proposes the optimal control method. Section 6 presents the simulation results and industrial applications in a certain zinc hydrometallurgy plant. Section 7 concludes the overall paper.

2. Description of the goethite process

2.1. Process description

The goethite process is an important part of the atmospheric direct leaching of zinc concentrates, which is introduced from the Outokumpu production technology. The simplified schematic of the process in a certain zinc hydrometallurgy plant in China is presented in Fig. 1.

It is observed that there are two primary operations in this procedure, the precipitation of iron and the solid-liquid separation. The iron precipitation procedure is comprised of four reactors, whose active volume is approximately 300 m^3. The inlet solution (zinc sulfate solution) flows from the #1 reactor to the #4 reactor, and its compositions are shown in Table 1.

The atmospheric and enriched oxygen gases are fed to the bottom of the reactors, and zinc oxide is added to four reactors. The temperature of solution is kept around at 353 K. In the solution, the ferrous ions are oxidized by oxygen and precipitated in the form of goethite. To guarantee the appropriate acid-base environment for the precipitation of iron, the surplus hydrogen ions are neutralized utilizing zinc oxide. After the iron is removed, the solution then flows into a thickener for the separation of the supernatant liquid from the precipitate. The goethite precipitate can be obtained from underflow of the thickener. Part of the underflow is circulated to the #1 reactor for the purpose seeding, which can promote the formation of goethite in the reactors. The technical requirement of the goethite process is that the mass concentration of ferrous ion in the final solution after the precipitation should be less than 1 g/L. However, from the view of production process, the final ferrous ion concentration is not the smaller the better. If the ferrous ions are removed to be below 0.3 g/L, the copper ions, which act as an important function in the next process (purification process), will be also removed. Therefore, in production the mass concentration of ferrous ion in the final solution is best controlled in [0.3 g/L, 0.8 g/L]. This is also an important index to assess the acceptability of the process.