ELSEVIER

Chemical Engineering Science 61 (2006) 229–245

Chemical Engineering Science

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Validation of a compartmental population balance model of an industrial leaching process: The Silgrain® process

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Received 1 June 2004; accepted 1 January 2005
Available online 27 June 2005

Abstract

Population balance (PB) models have become the most widely used tool for dynamic modeling of particulate processes. The application of PB models for prediction purposes is attracting significant interest. A parameter estimation and a quantitative validation of PB models should be carried out before the model can be applied for prediction. PB models are large-scale and nonlinear in the parameters. Moreover, the availability of measurements is typically limited, especially at industrial level, which makes the parameters poorly identifiable from experimental data. This paper shows how a systematic method for analyzing parameter sensitivity and collinearity among parameters, provides a subset of parameters that can easily be identified from the available data. A compartmental PB model of an industrial hydrometallurgical leaching plant is developed. Parameter identifiability of the model parameters is analyzed, and experimental data from the industrial plant are used to identify the corresponding subset of parameters and to verify some of the main assumptions of the model.

Keywords: Model validation; Parameter identification; Particulate processes; Population balance; Leaching; Hydrometallurgy

1. Introduction

The ideas of populations go back at least to Fisher's work in statistics, and the population balance was used e.g. by Flory in polymer growth modeling (Flory, 1953); see also Ramkrishna and Mahoney (2002) for some early applications. However, in its modern form, the population balance (PB) equation was developed in 1964 by two groups of researchers studying crystal nucleation and growth (Hulburt and Katz, 1964; Randolph, 1964). Since then, extensive research has been carried out on PB modeling of particulate processes. A good overview of PB modeling issues and applications is given in the review article by Ramkrishna (1985), and in the book by the same author (Ramkrishna, 2000).

Some reasons for the success of the PB approach are its ability to describe the wide variety of physical phenomena encountered in such processes and its parallelism with the well-known framework of conservation laws. The potential of PB models for use in diverse applications such as in process optimization, process control, state estimation, and operator training, is significant. However, in applications involving prediction, PB models should not be trusted until the realism of the model is verified, and the model predictions are validated quantitatively with experimental data from a real, industrial process. Moreover, PB models have many and different types of parameters (physical constants, birth and death rate parameters, dimensional parameters, etc.), and their a priori accuracy may vary considerably. This makes it necessary to carry out a parameter estimation procedure prior to the validation of the predictive power of the model.

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doi:10.1016/j.ces.2005.01.047

Despite the vast amount of references on PB modeling, there are relatively few publications focusing on parameter estimation and quantitative validation of PB models. In many instances, the model parameters are taken from the literature, and validation is limited to a qualitative comparison of the simulation results with experimental data from laboratory-scale units (Galán et al., 2002; Gerstlauer et al., 2002; Immanuel et al., 2002; Zeaiter et al., 2002). Since PB models are typically nonlinear in the parameters, the resulting parameter estimation problem is posed as a nonlinear optimization problem. As discussed in the review paper by Rawlings and Muske (1993), optimization schemes are intuitive and do not in general require model simplification, but careful and systematic experimental design and selection of the objective function should be performed. In the PB framework, both the weighted least-squares method (Alopaeus et al., 2002; Bhatia and Chakarborty, 1992; Chen et al., 1998; Dash and Rohani, 1993; Togkalidou et al., 2004), and the maximum likelihood method (Matthews et al., 1996; Miller and Rawlings, 1994; Hu et al., 2004) have been used. Occasionally, a parametric sensitivity study is carried out after model building and before comparing with experimental data, e.g. Immanuel et al. (2002) and Rawlings and Ray (1988). Systematic methods for parameter selection prior to parameter estimation have not been exploited to a great extent in the PB framework. An exception is a group of researchers working with bioreactors, who has developed a method that accounts for output sensitivities to parameter variations and collinearities between parameter (Li et al., 2004; Mhaskar et al., 2002).

Note that all the references above, except for Galán et al. (2002), use experimental data from laboratory-scale reactors. In contrast, we use data from an industrial plant. Prior to the industrial campaign we had obtained values for some of the model parameters in a laboratory setup (Dueñas Díez, 2004), but not all conditions of the real process can be reproduced in a laboratory reactor. The model parameters obtained in the laboratory are used as nominal values for the parameter estimation when using the data gathered at the plant. Running experiments in an industrial plant clearly poses more challenges than a laboratory reactor, such as: lack of instrumentation, limited amount of data, noisy data, limited input excitation, unavoidable disturbances from other pieces of equipment, etc. One can thus expect that a systematic parameter selection procedure is more relevant in an industrial case than in a laboratory case.

Fortunately, parameter identifiability and parameter estimation of large-scale mechanistic models has been widely studied in other areas of engineering, see e.g. (Bard, 1974; Beck and Arnold, 1977; Walter and Pronzato, 1997). We have chosen the systematic approach described in Brun et al. (2001, 2002). This method provides identifiability diagnosis for parameter subsets and is suited to large simulation models. Two identifiability measures are calculated. The first accounts for the sensitivity of model predictions to single parameters, and the second for the degree of collinearity among parameters for various parameter subsets. These two criteria differ from those used in Li et al. (2004) and Mhaskar et al. (2002). Moreover, the chosen approach seems to be better suited to dynamic data then the one in Li et al. (2004) which was mainly developed for steady-state analysis. The chosen parameter identifiability and estimation approach is illustrated with the PB model of an industrial leaching process called the Silgrain® process (Elkem ASA, Norway) for the production of Silicon (Si) from Ferrosilicon (FeSi).

The paper is organized as follows. Section 2 describes the PB model of the Silgrain® process. Section 3 introduces the systematic approach to parameter estimation and model validation of PB models. Section 4 discusses the application of the approach to the case under study. Finally, some conclusions are given in Section 5.

2. PB model of the Silgrain® process

2.1. Process description

The *Silgrain®* process is a hydrometallurgical leaching process where high-purity Si metal is produced by leaching lumps of FeSi in a hot acidic solution of ferric chloride ($FeCl_3$) and hydrochloric acid (HCl). More precisely, the particulate raw material is an alloy within the quaternary system Si–Fe–Al–Ca, with a Si content in the range 90–94%. The other three elements, i.e., Fe, Al, and Ca, are the impurities, and appear in diverse binary, ternary, and quaternary intermetallic phases. Depending on the exact impurity content and on the casting conditions, the alloy may contain different intermetallic phases such as $\alpha FeSi_2$, $\beta FeSi_2$, $CaSi_2$, $CaAl_2Si_{1.5}$, Caalsifier, etc. (Margarido et al., 1993, 1997). The phase composition of the raw material in the *Silgrain®* process is known and controlled (but cannot be revealed due to proprietary reasons). During leaching, the acid attacks the crystalline structure of the FeSi, selectively dissolving the intermetallic phases containing the impurities of Fe, Al, and Ca, while leaving the Si unattacked. The exact reaction mechanism for each soluble intermetallic phase is unknown, but leaching has proven to be well-represented by the following overall reduction-oxidation reactions (Aas, 1971):

$$\mathrm{Fe(s)} + 2\mathrm{FeCl_3(aq)} \longrightarrow 3\mathrm{FeCl_2(aq)}, \quad (1)$$

$$\mathrm{Fe(s)} + 2\mathrm{HCl(aq)} \longrightarrow \mathrm{FeCl_2(aq)} + \mathrm{H_2(g)}, \quad (2)$$

$$\mathrm{Al(s)} + 3\mathrm{FeCl_3(aq)} \longrightarrow \mathrm{AlCl_3(aq)} + 3\mathrm{FeCl_2(aq)}, \quad (3)$$

$$\mathrm{Al(s)} + 3\mathrm{HCl(aq)} \longrightarrow \mathrm{AlCl_3(aq)} + \tfrac{3}{2}\mathrm{H_2(g)}, \quad (4)$$

$$\mathrm{Ca(s)} + 2\mathrm{FeCl_3(aq)} \longrightarrow \mathrm{CaCl_2(aq)} + 2\mathrm{FeCl_2(aq)}, \quad (5)$$

$$\mathrm{Ca(s)} + 2\mathrm{HCl(aq)} \longrightarrow \mathrm{CaCl_2(aq)} + \mathrm{H_2(g)}. \quad (6)$$