



# Additive requirement ratio prediction using trend distribution features for hydrometallurgical purification processes



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## ABSTRACT

A purification process is to remove impurities through a series of reactors with additives. The theoretical calculated amount of additive does not fulfill actual requirements due to variations in the reaction environment. An additive requirement ratio is thus defined to measure the disparity between theoretical calculation and actual requirements. Considering the influence of the process underlying variations, a novel ratio prediction strategy, case-based prediction with trend distribution feature (CBP-TDF), is developed. In the strategy, the trend distribution features are firstly extracted to describe the underlying variations, and an improved case-based prediction algorithm is proposed where the similarity between these features is computed based on Kullback–Leibler divergence. The proposed strategy is applied to a copper removal process of zinc hydrometallurgy. The experiments indicate the accuracy of the ratio prediction, and the industrial application shows its effectiveness in the control of the purification process.

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## 1. Introduction

In the hydrometallurgical processes, undesirable metal ions in a leaching solution are often harmful to the industrial production (Laatikainen, Lahtinen, Laatikainen & Paatero, 2010). The excess metal ions reduce the production efficiencies of later processes, and also easily result in energy waste and downgrade in product quality. These impurities are removed in several removal stages, where the impurity ions are usually precipitated by using additives (Ahmed, El-Nadi & Daoud, 2011; Amin, El-Ashtouky & Abdelwahab, 2007), of purification process. For example, in zinc hydrometallurgy, impurities in leached zinc sulfate solution, mainly including of copper, cobalt, nickel and cadmium, are separately deposited in three stages by adding zinc powder (Sun, Gui, Wu, Wang & Yang, 2013); and impurity (silver) in copper hydrometallurgy is precipitated by copper powder in the purification process (Hietala & Hyvarinen, 2004). During these stages, the amounts of these additives must be determined exactly in the process control. Insufficient amounts of additives cannot decrease the impurities to the required level, while excess amounts not only waste the additive and, during some special purification processes, decrease the removal efficiency of the next process (Li, Gui, Teo, Zhu & Chai, 2012).

The additive amount is usually calculated by a mechanism-

based or semi-empirical model (named additive model in this paper) with many process variables, such as the concentration of the target impurity, the temperatures, the flow rates and the pH of the solution (Zhang, Yang, Zhu, Li & Gui, 2013; Xie et al., 2015; Sun, Gui, Wang & Yang, 2014). During some processes in the real world, however, part of the additive is leached with water (for example, in zinc purification), and the resultant precipitate decreased the activity of the additive (Singh, 1996). This scenario consumes more additive than the theoretical amount. During some purification processes, the additive consumption might also be lower than the theoretical amount (Näsi, 2004; Stole-Hansen, Wregget, Gowanlock & Thwaites, 1997; Kim, Kim, Park, Song & Jung, 2007). The impurities in these processes could also be deposited during side reactions, saving a portion of the additive. Therefore, the amount of additive actually consumed usually differs from the theoretical amount.

To measure the difference between the theoretical amount and actual consumption of additive, some researchers have defined various coefficients to adjust this amount. Stole-Hansen et al. (1997) defined a stoichiometric efficiency factor as the ratio between the actual additive amount and the theoretical amount calculated based on CSTR and stoichiometric efficiency. This factor was applied in a feed forward control strategy to lower the variability of the outlet copper concentration in the copper removal process. Kim et al. (2007) proposed a time series-based estimation method for the efficiency of the additive during a copper removal process. The steady state additive efficiency was defined as the ratio between the realistic actual and stoichiometric

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amounts of zinc powder. The efficiency, considered as a time series, was estimated using the Box–Jenkins method. The additive efficiencies could be used to evaluate the process condition. And those studies provide the inspiration and motivation for our proposed method of measuring the gaps between the theoretical and real requirements of the additive during a purification process. However, these efficiencies proposed in the previous researches were defined based on an additive model corresponding to a specific removal process (e.g. the stoichiometric or CSTR models for copper removal process of zinc purification). When the control system is improved or the additive model is altered, the additive efficiency could not accurately measure the differences between the theoretical amount and actual consumption of additive. And also the efficiency definitions are difficult to be applied directly used in other similar hydrometallurgical purification processes. Therefore, an additive requirement ratio (ARR) is defined for the hydrometallurgical purification processes in this paper.

ARR varies with the process condition. The present value of ARR could not appropriately amend the theoretical additive amount which is set for the future removal work. ARR prediction is thus essential in the removal process control. ARR reveals the efficiency of the reactant. This value depends on the current reaction condition, and it is cumulatively affected by historical variables especially in the hydrometallurgical purification processes. Therefore, the historical trends in the process parameters must be considered when predicting ARR. Trend analysis is a useful approach for extracting cumulative information from numerical data and the present range in the variations, and it can also reveal underlying changes of the process (Villegz, Rosén, Anctil, Duchesne & Vanrolleghema, 2013; Howell, Bevan & Burr, 2013; Demirkiran, Ekmekyapar, Künkül & Baysar, 2007; Gamero, Meléndez & Colomer, 2014). To extract trend information from the process variables, a novel process information extraction method based on quantitative trend analysis is proposed here. In this method, a historical variable is cut into a group of time segments and classified into several trend sets; then, the distribution information is extracted from these trends and treated as the trend distribution feature (TDF) of the process.

During ARR prediction process, a prediction algorithm plays an essential role. Recently, various algorithms have been used to solve this type of problem, such as neural networks (Ghavipour, Ghavipour, Chitsazan, Najibi & Ghidary, 2013; Yang, Gui, Kong & Wang, 2009; Iliyas, Elshafei, Habib & Adeniran, 2013), support vector regressions (Xi, Poo & Chou, 2007; Han, Liu, Zhao & Wang, 2012),

partial least squares (Wang, Jang, Wong, Shieh & Wu, 2013; Vanlaer, Gins & Impe, 2013), autoregressive integrated moving averages (Zhang, Teng & Zhang, 2010; Pellegrini, Ruiz & Espasa, 2011), case based reasoning (CBR) (Qi, Hua, Peng, Chai & Ren, 2013; Xing, Ding, Chai, Puya & Wang, 2012; Chang, Fan & Lin, 2011) etc. In this paper, CBR is chosen for predicting ARR for following reasons: (1) it is relatively easy to set up a knowledge base; (2) CBR can be used in the problem domains that are not well understood; (3) in particular, CBR is easy to understand and has low computation cost, which is helpful for future maintenance and real-time process control (Chou, 2009).

The remainder of this paper is organized as follows. Section 2 illustrates the definition of ARR for a hydrometallurgical purification process. Section 3 describes the proposed case-based prediction strategy, which is called case-based prediction with trend distribution features (CBP-TDF) for ARR. The prediction strategy consists of the trend classification, the trend distribution extraction and the case similarity calculation. In Section 4, a real copper removal process is described, and a series of experiments are performed to verify the prediction accuracy of CBP-TDF. To verify the effectiveness of the proposed approach on improving the removal process control, ARR prediction is planted into two different control strategies to amend the theoretical additive amounts. The simulation results of these improved control strategies, using the samples from the copper removal process, are also afforded. Subsequently, the application of the proposed ARR to this process and the results are also discussed. Finally, our conclusions are given in Section 5.

## 2. Additive requirement ratio for removal stages

A hydrometallurgical purification process usually occurs through a series of continuous stirred tank reactors (CSTRs). During the conventional process, the solution flows from the first reactor to the last one where the impurities are removed into a required concentration, as shown in Fig. 1(a). Due to the variety and diversity of the sources of concentrate, the concentrations of the ionic metals vary constantly, and the reaction conditions are more complex than expected. Therefore, the additive amount is imprecisely determined. This situation is also evident in some improved purification process where the underflow is recycled to the first reactor, as shown in Fig. 1(b). The low precision of the additive amount determination occurs for two major reasons.

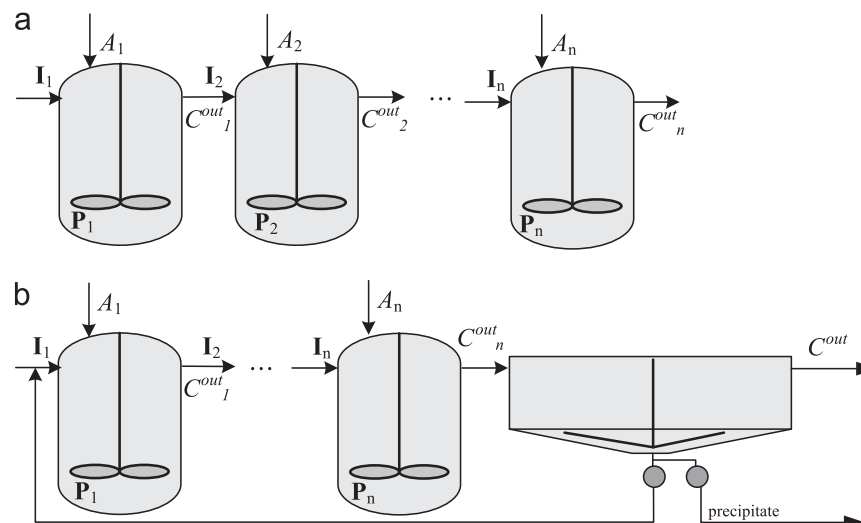


Fig. 1. The hydrometallurgical purification processes: (a) a conventional process, (b) an improved process with recycling underflow.

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