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## Pollution prevention and control measures for the bottom blowing furnace of a lead-smelting process, based on a mathematical model and simulation

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#### ABSTRACT

One of the outstanding environmental protection problems of lead smelting enterprise at present is that the large amount of generation and emissions of flue gas and dust contaminated with heavy metals, particularly uncontrolled emissions (such as dissipation of dust). In order to solve the mentioned problem, this article takes the lead smelting process as the research object, and develops a mathematical model to simulate the production process of lead smelting. The model, which is commonly used in the production practice of metallurgical industry, is improved and verified in this article to simulate the output of products and pollutants under different operational conditions. The developed model was applied to simulate a bottom blowing furnace in a lead-smelting plant with 1 t of input materials fed in at a continuous normal production level, and verified by measured data from the actual production. Further simulation was carried out to analyze the influences of different reaction parameters, such as the composition of input materials, smelting temperature, and oxygen enrichment (input oxygen amount) on the output constituents, and Pb behavior among the various phases, particularly in flue gas. The results show that when the total content of Pb and S in the input materials, and other operational parameters, are constant, a Pb content of 52-62% in the input materials will be beneficial for reducing the Pb amount in flue gas as well as increasing the output amount of lead bullion. When the composition (species and content) of input materials and oxygen enrichment are constant, a lower temperature is more advantageous to reduce the Pb amount in the flue gas. When the composition (species and content) of the input materials and the smelting temperature are constant, the Pb amount of the gas and the flue gas will be reduced significantly if the input oxygen amount is larger than 90 m<sup>3</sup>/t-input materials. Finally, the most suitable operational scheme for the higher production efficiency and fewer pollutant generation amounts are discussed and suggested.

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#### 1. Introduction

Cleaner production, as it known, mainly refers to the improvement of production process of enterprises, such as raw materials, process control, technical level, staff skills and management level and so on. Cleaner production is quite often used as a diagnose tool for pollution prevention and control of a factory. When we dissect a factory from the perspective of cleaner production, lots of improvement measures could be put forwarded to improve the entire efficiency of the production system, including fewer

\* Corresponding author. E-mail address: qiaoqi@craes.org.cn (Q. Qiao). pollutants generation and higher product output. Therefore, the practice of cleaner production cannot be separated from the production process, and should be more focused on cleaner and environmentally friendly technology from the perspective of process improvement.

Due to the large amount of generation of heavy metals contained flue gas and dust in the production process of non-ferrous metal smelting industry (including lead smelting), pollution prevention and control of non-ferrous metal smelting industry has a great significance on environmental risks and human health. Furthermore, control of lead in flue gas plays a vital role for a lead smelting process, in both of efficiency of resource utilization and environmental protection. In order to prevent dissipation of the smoke and dust, with their heavy-metal content, adjustment of







Abbreviation list		R	the Gas constant; $R = 8.314 \text{ J/(mol K)}$
SKS	an abbreviation of "Shui kou shan", a type of lead- smelting process	$x_j$	the mole fraction (mole number proportion, %) of component <i>j</i> in the subordinate phase
BBF	an abbreviation of "bottom blowing furnace"	$\gamma_i$	the activity coefficient of component <i>j</i>
ME Model an abbreviation of "multiphase equilibrium" Model		m	the substance phase, $m = 1,2,3 \dots k$
Т	temperature of the given system	$S_m$	the number of components in phase <i>m</i>
Р	pressure of the given system	n <sub>i.i</sub>	the atomic number of element <i>i</i> in component <i>j</i>
S	the number of components in the given system	$b_i$	the total mole number of element <i>i</i> in the given system
k	the number of substance phases in the given system	C	the total elements number in the given system
G	Gibbs free energy of the given system	$\lambda_i$	the element potential of element <i>i</i>
Ni	the total mole number of component j	Q	a constructed function
$G_j$	the sub-Gibbs function of component $j$	$\widetilde{N}_{(m)}$	the total mole number of component $j$ in phase $m$

influencing factors (operational conditions) to a proper level, or taking some cleaner production measures, would be very helpful.

The smelting process of non-ferrous metals usually can be viewed as a high temperature-multiphase-multicomponent equilibrium system that follows thermodynamic principles and the law of conservation of mass. Based on these principles, mathematical models of smelting processes have been developed to simulate and improve the reaction processes, such as speeding up the reaction rate or increasing the output product amounts. Usually, the methods for transforming lead compounds (either naturally occurring or artificial) into the elemental substance of lead can be divided into two basic categories: pyrometallurgy and hydrometallurgy. Pyrometallurgy is the most popular of these two methods, and since the pyrometallurgical smelting process is essentially a complex, high-temperature, multiphase, and multicomponent system, a mathematical model of high-temperature/multiphase/ multicomponent equilibrium, based on thermodynamic principles and the law of conservation of mass (hereinafter referred to as a multiphase equilibrium mathematical model, or ME Model), was developed in the early 1970s. This mathematical model could help us to better understand the complicated reactions occurring in the smelting process. Japanese scholar Goto developed a model to simulate a flash smelting process (Goto, 1975). Since then, mathematical models and computer simulations of pyrometallurgy (especially flash smelting processes) have been established by many researchers. The reports of mathematical models of pyrometallurgy published so far have mainly been focused on copper smelting (Kim and Sohn, 1996; Wang et al., 2013; Gui et al., 2007; Liu et al., 2014). These models are designed to simulate actual production conditions, in order to calculate a group of best operational parameters aimed at improving production efficiency. Generally, increasing the output copper amount or Cu content in products has been given priority in these research works.

In fact, however, mathematical models can provide not only production amounts, but also the amounts of by-products such as gases and other substances contained in the compounds, or traces of precious and rare elements (Shuva et al., 2016). Therefore the mathematical model of pyrometallurgy can also be applied to simulate operational parameters influencing the generation amount of by-products, including heavy metals in gas. A better understanding of the behavior of by-products such as heavy metals, particularly Pb, could not only improve production efficiency but also provide measures for eliminating flue gas and dust.

However, mathematical models of lead-smelting processes are limited, and by-products such as heavy metals in flue gas have usually been neglected in those published reports. Besides, ME model is used only in order to get more material output (products), or products of higher concentration in the past. Is that more output products represents a higher efficiency of the whole production system, which requires deep thinking. In pursuit of higher production of a product, we should also pay attention to the pollutants generated at the same time. The ultimate purpose of this article is to help the factory who uses this technology could increase the control of pollutant generation and emission while they concentrate on the production control. Thereby the pollutants generation, particularly the dust and flue gas contaminated with heavy metals could be prevented or eliminated from the very beginning.

#### 2. Modeling and analytical method

#### 2.1. Solving method for multiphase equilibrium model

The two most popular methods for calculating a multiphase equilibrium model in chemistry are: a method based on the equilibrium constant (K-value method) and a method based on Gibbs free-energy minimization. The method based on the equilibrium constant was put forward by Sanderson and Chien in 1973 (Sanderson and Chien, 1973). An algorithm of Newton Raphson is usually employed to calculate the electric-neutral equations, the phase equilibrium and the chemical equilibrium equations, in this method. Although the algorithm converges rapidly and is easy to program for a computer, it needs to be given independent reaction equations in advance, and there are too many equations to be solved, making it not very suitable for a complex system for which some phases and reactions are unknown.

The method based on the Gibbs free-energy minimization was put forward by White (White et al., 1958) for the first time in 1958. The Gibbs free-energy minimization principle, which can be expressed as the total Gibbs free energy of a system, will be at its minimum when the system reaches chemical equilibrium. This principle can be applied to thermodynamic phase equilibrium systems as well. The phase equilibrium calculation problem therefore could be transformed into a nonlinear programming problem with constraints, i.e. an optimization problem, with this method. The mole number or reaction activity of each product at equilibrium is set as the variable for calculating the total Gibbs free energy of the system, and then compositions of each phase can be obtained by solving a mathematical optimization problem based on the Gibbs free-energy minimization principle.

The original approach to solving the Gibbs free energy (Gibbs function) of a closed system was to view the Gibbs function as quadratic function, or approximate quadratic function, and build a Lagrange function such as the RAND method (Dluzniewsli and Adler, 1972), the Wolfe method of quadratic programming (Gautam and

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