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Process optimization for vacuum distillation of Sn–Sb alloy by response surface methodology

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ABSTRACT

Based on the molecular interaction volume model (MIVM), the vapor–liquid phase equilibrium of Sn–Sb alloy was calculated, which was used to predict the element distribution of Sn–Sb alloy between vapor and liquid phase during vacuum distillation. A central composite design (CCD) was used to optimize the process parameters influencing the content of Sn in liquid phase and the direct yield of Sn. The studied parameters were distillation temperature, feeding materials and soaking time. Two quadratic mathematical model equations were derived for predicting the content of Sn in liquid phase and the direct yield of Sn. The analysis of variance (ANOVA) shown that distillation temperature was the most significant factor affecting the separation of Sn–Sb alloy. In the process optimization, while the direct yield of Sn equal to 92%, the maximum content of Sn in liquid phase should be 99.66 wt.% under the conditions of 1531 K, 137 g and 46 min. The confirmation test values of 91.22% and 99.43 wt.% were fair agreement with the predicted data, which demonstrated that these models were very good and can be used for parameter optimization in vacuum distillation.

1. Introduction

Tin is an indispensable metal, the major uses of which are in tin plate, solder and specific alloys. More recently, the increasing demand for tin metal and the high price of it have encouraged the purification of crude tin. Normally, the crude tin produced with both crude ores and secondary metals, contains Pb, Sb, Bi, etc. So, it is imperative for tin smelters to find a convenient and economic method to separate these metals [1]. Conventional refining techniques including pyrometallurgical [2] and electrowinning [3–5] have been generally used as the methods of purification and refining of crude tin. However, there still exist some deficiencies, such as low metal recovery, high energy consumption, evident environment pollution, etc. Vacuum refining of crude tin, in contrast to above methods, is more efficient, both in terms of the high recovery ratio of Sn and low operation costs [6,7]. Previously, JIANG Guang-you et al. [8] verified that the separation of high antimony crude tin by vacuum distillation can be easily realized. Kunming University of Science and Technology in China have been making efforts to separate Sn-based alloys by vacuum distillation technology for years. Sn–Sb alloy is one of the most important types of Sn-based alloys. However, its separation has not been investigated and reported.

It is generally known that the key process parameters affecting the separation of Sn–Sb alloy during vacuum distillation normally are distillation temperature, feeding materials, soaking time and ambient pressure. From the industrial point of view, it is essential to find out the optimum process parameters. However, the traditional single-factor experiment is time-consuming and laborious. Moreover, it is easy to ignore the interaction among the factors and cannot realize the process optimization. While response surface methodology (RSM) can well overcome above shortcomings, evaluate the effects of factors and their interactions and determine the best level range [9].

The purpose of this work was to obtain optimum process parameters of Sn–Sb alloy during vacuum distillation. In our previous study, the effects of distillation temperature, feeding materials, soaking time and ambient pressure have been studied. It was found that the ambient pressure has little or no effect on the separation of Sn–Sb alloy when it less than 30 Pa. Combining with industry

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practices (ambient pressure normally ranging from 10 to 30 Pa), hence, the effects of distillation temperature, feeding materials and soaking time on the content of Sn in liquid phase and the direct yield of Sn were investigated and optimized by using the RSM in this paper.

2. Methods

2.1. Model predictions

The thermodynamic property is very important in metallurgical process. Experimental thermodynamic study, however, is time-consuming, for a great number of measurements have to be taken. Therefore, theoretical prediction is such an effective and significant approach to obtain thermodynamic properties of an alloy [10]. HongWei Yang et al. [11] applied the molecular interaction volume model (MIVM) to Pb-based alloys separation in vacuum distillation. Then LingXin Kong et al. [1,12] extended the MIVM to Sn–Pb–Sb ternary alloy. Their researches show that MIVM is reliable and convenient for vacuum distillation. Therefore, the MIVM was employed to predict the activity coefficients of Sn–Sb alloy over the temperature range 1073–1673 K in this study.

Tao [13] developed MIVM from statistical thermodynamics, requiring no assumptions and using fewer fitting parameters, and predicted thermodynamic properties of some alloys [14,15]. These activity coefficients γ_{Sn}, γ_{Sb} of Sn and Sb in Sn–Sb alloy can be obtained with MIVM using experimental data γ_{Sn}^{∞}, γ_{Sb}^{∞} and Newton–Raphson methodology. The values of these physical parameters for Sn–Sb alloy of interest in this work are available in Refs. [16,17].

The activity coefficients calculated with MIVM are listed in Table 1, as well as the experimental data [18]. Table 1 shows that the calculated values are nicely consistent with the experimental data, which indicates that MIVM is reliable.

2.1.1. Separation coefficients

Y.N. Dai [19] conducted the separation coefficient, β, to determine whether two elements can be separated from each other by vacuum distillation according to thermodynamic equilibrium theory. The separation coefficient is presented as follows:

$$\beta = \left(\frac{\gamma_i}{\gamma_j}\right) \cdot \left(\frac{P_i^*}{P_j^*}\right) \tag{1}$$

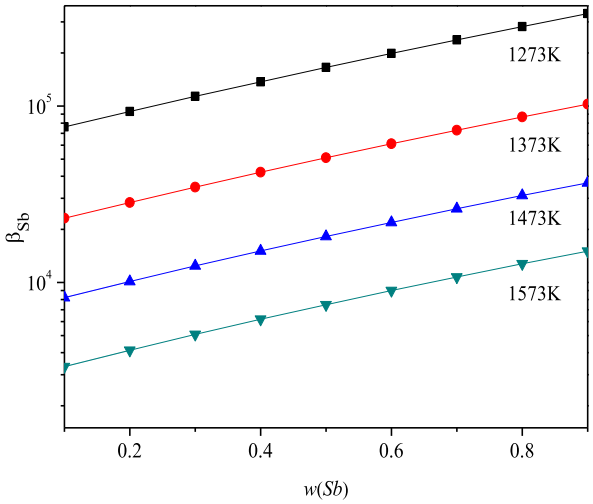

Fig. 1. The separation coefficients of Sn–Sb alloy at different temperatures, calculated by the MIVM.

where γ_i and γ_j are the activity coefficients of i and j components, respectively;P_i^* and P_j^* are the saturated vapor pressures of i and j in pure state, respectively.

After we substitute activity coefficients γ_i and γ_j at different temperatures into Eq. (1), these separation coefficients can be easily calculated as shown in Fig. 1. It shows that the content of Sb in gas is thousands to ten thousands higher than those in liquid. So, Sb can be concentrated and purified in gas phase by vacuum distillation.

2.1.2. Vapor–liquid phase equilibrium

The element distribution of Sn–Sb alloy between vapor and liquid phase was investigated by calculating the vapor–liquid equilibrium [19] in this work. The mass fraction of i in vapor phase can be expressed as:

$$\omega_{i,g} = \left[1 + \frac{\omega_{j,l}}{\beta \cdot \omega_{i,l}}\right]^{-1} = \left[1 + \left(\frac{\omega_{j,l}}{\omega_{i,l}}\right) \cdot \left(\frac{\gamma_j}{\gamma_i}\right) \cdot \left(\frac{p_j^*}{p_i^*}\right)\right]^{-1} \tag{2}$$

where ω, γ, p and β are mass fraction, activity coefficient, saturated vapor pressure and separating coefficient, respectively. The saturated vapor pressures of pure Sn and Sb at different temperatures are available in Ref. [19].

Table 1
The experimental and calculated activity coefficients of Sn–Sb alloy system.

T/K		γ	X_{Sn}											
			0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	0.99	0.999	0.9999
Experiment	905	γ_{sn}	0.486	0.566	0.647	0.726	0.800	0.867	0.923	0.965	0.991	–	–	–
		γ_{sb}	0.991	0.965	0.923	0.867	0.800	0.726	0.647	0.566	0.486	–	–	–
Calculated by MIVM	905	γ_{sn}	0.487	0.567	0.648	0.727	0.800	0.867	0.923	0.965	0.991	–	–	
		γ_{sb}	0.991	0.965	0.923	0.867	0.800	0.727	0.648	0.567	0.487	–		
	1073	γ_{sn}	0.484	0.561	0.640	0.718	0.793	0.861	0.918	0.962	0.990	0.999	1.000	1.000
		γ_{sb}	0.991	0.966	0.924	0.869	0.801	0.725	0.643	0.558	0.474	0.400	0.380	0.380
	1173	γ_{sn}	0.483	0.559	0.637	0.715	0.789	0.858	0.916	0.961	0.990	0.999	1.000	1.000
		γ_{sb}	0.992	0.966	0.925	0.869	0.801	0.724	0.640	0.554	0.469	0.400	0.377	0.377
	1273	γ_{sn}	0.481	0.557	0.634	0.711	0.786	0.855	0.914	0.960	0.990	0.999	1.000	1.000
		γ_{sb}	0.992	0.967	0.926	0.870	0.801	0.723	0.639	0.551	0.464	0.390	0.372	0.372
	1373	γ_{sn}	0.481	0.555	0.631	0.709	0.783	0.853	0.912	0.959	0.989	0.999	1.000	1.000
		γ_{sb}	0.992	0.967	0.926	0.870	0.801	0.723	0.637	0.548	0.460	0.380	0.367	0.367
	1473	γ_{sn}	0.480	0.553	0.629	0.706	0.781	0.851	0.911	0.958	0.989	0.999	1.000	1.000
		γ_{sb}	0.992	0.967	0.926	0.871	0.802	0.722	0.636	0.546	0.456	0.380	0.362	0.362
	1573	γ_{sn}	0.479	0.552	0.627	0.704	0.779	0.849	0.910	0.958	0.989	0.999	1.000	1.000
		γ_{sb}	0.992	0.968	0.927	0.871	0.802	0.722	0.635	0.544	0.453	0.370	0.359	0.359
	1673	γ_{sn}	0.478	0.551	0.626	0.702	0.777	0.847	0.909	0.957	0.989	0.999	1.000	1.000
		γ_{sb}	0.992	0.968	0.927	0.871	0.802	0.722	0.634	0.542	0.450	0.370	0.355	0.355