



## Experimental investigation and modelling of phase equilibria for the Ag–Cu–Pb system in vacuum distillation



WenLong Jiang<sup>c</sup>, Cheng Zhang<sup>b</sup>, Na Xu<sup>d</sup>, Bin Yang<sup>c</sup>, BaoQiang Xu<sup>c</sup>, DaChun Liu<sup>c</sup>, HongWei Yang<sup>a, b, c, \*</sup>

<sup>a</sup> The State Key Laboratory of Complex Nonferrous Metal Resources Clean Utilization, Kunming University of Science and Technology, Kunming, 650093, PR China

<sup>b</sup> Faculty of Metallurgical and Energy Engineering, Kunming University of Science and Technology, Kunming, 650093, PR China

<sup>c</sup> National Engineering Laboratory for Vacuum Metallurgy, Kunming University of Science and Technology, Kunming, 650093, PR China

<sup>d</sup> Kunming Metallurgical Research Institute, Kunming, 650031, PR China

### ARTICLE INFO

#### Article history:

Received 9 November 2015

Received in revised form

8 February 2016

Accepted 14 February 2016

Available online 16 February 2016

#### Keywords:

Vacuum metallurgy

VLE phase diagram

Thermodynamic properties

Wilson equation

### ABSTRACT

In this work, vapor–liquid equilibria (VLE) of the Ag–Cu–Pb system were experimentally investigated in vacuum distillation. The experimental results show that the content of Pb in liquid phase is 0.005 (mole fraction) under the experimental condition of 1173 K, 90 min and 10 Pa. The amount of Pb in volatiles increased to 0.9911 (mole fraction). It indicates that Pb can be satisfactorily removed from Ag–Cu alloy. With the Wilson equation, the partial Gibbs free energies of Pb in liquid Ag–Cu–Pb alloys were calculated, and the predicted values agree well with the experimental data, which shown that the interaction parameters are reliable. Using binary data only, the VLE for the Ag–Pb and Ag–Cu–Pb systems under vacuum condition were obtained. The calculation results demonstrate that this approach can lead to accurate VLE data predictions for the binary and ternary systems based on the Wilson equation.

© 2016 Elsevier B.V. All rights reserved.

### 1. Introduction

Vacuum distillation has been successfully used in separating various elements from nonferrous alloys and the area of its application is being extended rapidly. It has the advantages of high metal recovery, low impurities in recovered metal, simplified flow sheet, good environmental protection, low operation costs, and so on [1,2]. Reliable vapor–liquid equilibria (VLE) data are necessary for design and operation of separation processes for crude metals in vacuum distillation. Due to experimental difficulties in accurate measurement of VLE for alloy systems, theoretical predicting is a significant and effective method to obtain thermodynamic properties, especially for multicomponent systems [3,4].

Lead-zinc ores contain various valuable metals, including Cu, Ag and other trace metal impurities. Crude copper usually contains lead as an impurity, the conventional separating method cannot be used when the lead is high. In our previous work, vacuum

distillation experiments for Ag–Pb and Cu–Pb alloys were carried out [5,6]. Small-scale vacuum distillation experiments to purify the Pb contaminated alloys have been investigated which got good separation effects. The Phase diagrams for vapor–liquid phase of the binary systems were calculated using thermodynamic models. Good results were obtained for these systems.

In this paper, vacuum distillation of high-lead crude copper was investigated with vapor pressures at 10 Pa. The vapor–liquid phase equilibria of the Ag–Cu–Pb system were experimentally investigated in vacuum distillation. The VLE phase diagrams such as temperature-composition ( $T-x$ ) curves for Ag–Cu–Pb ternary system are obtained by using the VLE calculation and the Wilson equation under the experimental condition. The Wilson equation based on the concept of local composition which provides a very good representation of non-ideal mixtures [7]. A particular advantage of the Wilson equation is that it can be generalized to multicomponent mixtures without any additional parameters.

The objective of this work is to show application of the Wilson equation to the calculation of multicomponent vapor–liquid equilibria based only on data for their binary mixtures. The main value of the calculation method is that the data required for its application become excessive or too difficult to obtain. It provides

\* Corresponding author. The State Key Laboratory of Complex Nonferrous Metal Resources Clean Utilization, Kunming University of Science and Technology, Kunming, 650093, PR China.

E-mail address: [hongwei325@gmail.com](mailto:hongwei325@gmail.com) (H. Yang).

an intuitive way to analyze and predict the VLE data of alloys in vacuum distillation, especially for the process of ordinary distillation, stage batch distillation, or even pressure swing distillation in the vacuum furnace.

## 2. Experimental procedure

Samples 1# and 2# of the Ag–Cu–Pb ternary system with 100 g each for the experiment were prepared using pure Ag (99.99 wt. %), Cu (99.9 wt. %), and Pb (99.99 wt. %) obtained from Yunnan Tin company in China, which the contents of Pb in the samples are 0.0957 and 0.9412 (mole fraction), respectively. The details of materials and samples are summarized in Tables 1 and 2. The prepared metal pieces were then melted in an induction furnace under high purity argon atmosphere.

Small-scale vacuum distillation experiments for Ag–Cu–Pb alloy were carried out in the vertical vacuum furnace in this work and the internal structure schematic diagram is shown in Fig. 1. Vacuum degree in the furnace was at the range of 10 Pa during the experiment. The maximum furnace temperature can be heated up to 1700 K. The absolute vacuum pressure reached less than 1 Pa before heating by operation of pump. The vacuum chamber was evacuated to about 5 Pa by Rotary vane vacuum pump. Temperature was measured with Pt-100 probes connecting to a digital temperature meter (ANTHONE LU-900 M) with an uncertainty of  $\pm 0.01$  K. The pressure has been measured with a stand McLeod Gage which is accurate to within 1% at pressures below 300 Pa. The composition of the liquid samples was determined chemical analysis methods.

At each experiment, 100 g Ag–Cu–Pb alloy was put into the graphite crucible and placed inside the distillation section of the vacuum furnace. The vacuum pump started to work and vacuum furnace started to heat when the vacuum degree in the furnace was equal or less than 10 Pa. When temperature rise to a preset temperature, the temperature could be kept constant (in a very small range). Lead in liquid alloy will evaporate from the melt and solidify rapidly on the condenser. Melting temperatures of each experiment during this investigation were 1073 K, 1123 K, 1173 K, 1223 K, 1273 K, and 1323 K, respectively. The composition of distillation product started to approach to a stable level when the time over 60 min under 10 Pa. Therefore, 90 min was selected as the final distillation time in this work which can make sure the system approach to the phase equilibrium sufficiently. The samples were collected from the volatile and residue and then were melted respectively to determine the average components of the samples.

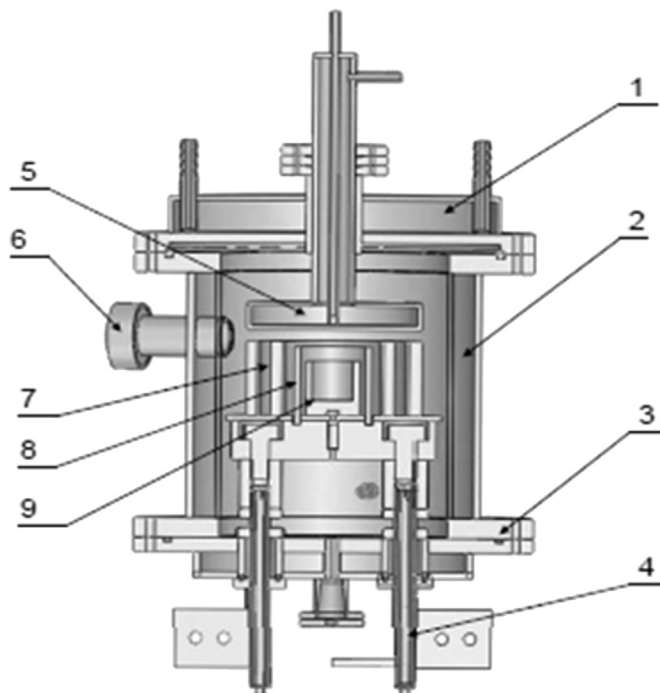
Experiments have been carried out in the temperature range 1100 K–1500 K at intervals of 50 K. The time for system to achieve equilibrium state was determined by distilling 60, 90, and 120 ... 600 min. In this process, tin and antimony in liquid alloy would evaporate from the melt and condensed rapidly on the condenser when the temperature rose to a certain degree. After the system achieved a state of equilibrium, the quality of liquid and vapor phase remained unchanged within a small range, respectively. The time for system to achieve equilibrium state increases gradually with the decreasing distillation temperature. Finally, the distillation time was set from 150 min at 1500K–420 min at 1100 K

**Table 1**  
Material description.

Chemical name	Source	Mass fraction purity	Purification method
Silver	Yunnan Tin company	> 99.99%	None
Copper	Yunnan Tin company	> 99.90%	None
Lead	Yunnan Tin company	> 99.99%	None

**Table 2**  
Chemical composition of Ag–Cu–Pb alloys with different composition.

Sample	$x_{Ag}$	$x_{Cu}$	$x_{Pb}$
1#	0.1790	0.7253	0.0957
2#	0.0531	0.0057	0.9412



**Fig. 1.** Schematic diagram of vertical vacuum furnace: 1 furnace lid; 2 furnace body; 3 furnace bottom; 4 electrode; 5 cold plate; 6 observation door; 7 heat holding cover; 8 heating unit; 9 graphite evaporator.

(maintained long enough time to ensure that the system was stable). The samples of the vapor and liquid phase were obtained from volatiles and residues, respectively. The contents of Ag and Cu in liquid phase were measured analyzed with a gas chromatograph (GC) (SP-2100, supplied by Kunming Metallurgical Research Institute). Since the contents of Ag and Cu in vapor phase are very small, the samples were accurately determined by atomic absorption spectrometry (PinAAcle 900, supplied by Kunming Metallurgical Research Institute). The contents of Pb in liquid phase and vapor phase are obtained by solving mass balance equation accounting for the vapor–liquid equilibrium, respectively. At the experimental conditions in this work, the estimated experimental uncertainty in  $u(x)$  and  $u(y)$  of these apparatus is 0.0002.

## 3. Thermodynamic models

### 3.1. VLE calculation

At equilibrium the fugacity of a component in the liquid equals the fugacity of that component in the vapor [8,9].

$$\bar{f}_i^L(x_i, T, P) = \bar{f}_i^V(y_i, T, P) \quad (1)$$

where  $\bar{f}_i^L$  and  $\bar{f}_i^V$  represent the fugacity of species  $i$  in the liquid phase and vapor phase, respectively, in terms of temperature, pressure and composition;  $x_i$  and  $y_i$  are the mole fraction of species  $i$  in the liquid phase and vapor phase, respectively;  $T$  and  $P$  are the

Download English Version:

<https://daneshyari.com/en/article/200803>

Download Persian Version:

<https://daneshyari.com/article/200803>

[Daneshyari.com](https://daneshyari.com)