

# Experimental investigation and calculation of vapor–liquid equilibria for Cu–Pb binary alloy in vacuum distillation



Cheng Zhang<sup>b</sup>, WenLong Jiang<sup>c</sup>, Bin Yang<sup>c</sup>, DaChun Liu<sup>c</sup>, BaoQiang Xu<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

## ARTICLE INFO

### Article history:

Received 18 April 2015

Received in revised form 18 July 2015

Accepted 23 July 2015

Available online 29 July 2015

### Keywords:

Vacuum distillation

VLE phase diagram

Cu–Pb alloy

Wilson equation

Thermodynamic properties

## ABSTRACT

In this study, vacuum distillation experiments of high-lead crude copper were performed under the vapor pressure 5 Pa. The content of lead in crude copper was reduced from 15% to less than 0.01% in a single stage distillation process with residual vapor pressures of below 10 Pa, distillation temperatures of 1423 K and distillation times of 60 min. The vapor–liquid equilibrium (VLE) phase diagrams of Cu–Pb binary alloy in vacuum distillation were calculated using the Wilson equation. Thermodynamic experimental data taken from literature were used for calculating Wilson equation temperature dependent interaction parameters. The calculations give a satisfactory accuracy with the experimental data for separation of Cu–Pb alloy in vacuum distillation. The results indicate that VLE phase diagrams under vacuum obtained by this method are reliable for predicting the process of vacuum distillation for Cu–Pb alloy. The VLE phase diagrams of alloys will have significant benefits for the industrial production of vacuum metallurgy especially for the process of multistage distillation in the vacuum furnace.

© 2015 Elsevier B.V. All rights reserved.

## 1. Introduction

The use of vacuum distillation for separating various elements from nonferrous alloys have been very successful and is expanding rapidly, largely due to its ability to achieve high metal recovery, low impurities in recovered metal, simplified flow sheet, good environmental protection, low operation costs and simple equipment [1,2]. Small-scale vacuum distillation experiments to purify the Pb contaminated alloys have been investigated in previous work and have reported good separation effects [3–5].

There is an increasing need to consider phase diagrams for vapor–liquid phase with the development of vacuum metallurgy. In our previous works [6,7], we only calculated the separation coefficients and vapor–liquid equilibrium composition of Pb-based alloys in vacuum distillation. However, the vapor–liquid equilibrium composition curve cannot provide an intuitive and convenient way to analyze the product component dependence of temperature or pressure in the design of vacuum distillation experiment. A

firm knowledge of the VLE relationships is a prerequisite to predict the process of vacuum distillation [8,9].

Crude copper usually contains lead as an impurity, the conventional separation method cannot be used when the lead is high. In this work, vacuum distillation of high-lead crude copper was investigated with vapor pressures ranging from 5 to 10 Pa. The VLE phase diagrams such as temperature–composition ( $T$ - $x$ ) and pressure–composition ( $P$ - $x$ ) curves of Cu–Pb alloy under vacuum are obtained based on the VLE calculation and the Wilson equation. The VLE phase diagrams can provide an intuitive way to predict the VLE relationships of alloy system under vacuum distillation continuously. The present work is available for obtaining the VLE phase diagrams of alloys which will bring an efficient and reliable way to analyze and predict the VLE data of alloys in vacuum distillation, especially for the process of multistage distillation in the vacuum furnace.

## 2. Experimental procedure

The Cu–Pb alloy for the experiment was prepared from high purity metals using Pb and Cu with a purity of 99.99% and the content of Pb in Cu–Pb alloy is 15%. The information of experimental sample for the Cu–Pb alloy is listed in Table 1.

\* Corresponding author at: The State Key Laboratory of Complex Nonferrous Metal Resources Clean Utilization, Kunming University of Science and Technology, Kunming 650093, PR China. Tel.: +86 13987157570.

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

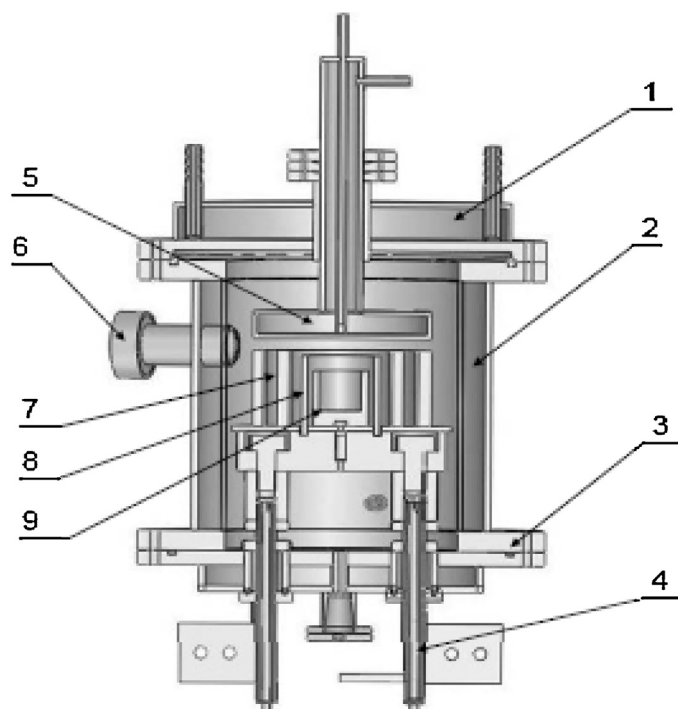
**Table 1**

The information of experimental sample for the Cu–Pb alloy.

Chemical name	Source	Initial mole fraction purity	Purification method	Final mole fraction purity	Analysis method
Copper	Yunnan tin company	0.85	Vacuum distillation	0.9982	Gas–liquid chromatography
Lead		0.15		0.0016	Atomic absorption spectroscopy

Small-scale vacuum distillation experiments for 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 were at the range of 5 ~ 10 Pa during the experiment.

In the furnace, 50 g Cu–Pb alloy was placed in graphite crucible to be heated and melted after the vacuum degree reached to 5 Pa. With the temperature rising to a certain degree, lead and copper in liquid alloy will evaporate from the melt and solidified rapidly on the condenser. Melting temperatures of each experiment during this investigation were 1223 K, 1273 K, 1323 K, 1373 K and 1423 K, respectively. The relationship between distillation time and composition of distillation product has been investigated in our previous works [10] and the composition of distillation product started to approach a stable level when the time exceeds 30 minutes under 10 ~ 25 Pa and 1373 K. In other words, the stable level of composition of distillation product is the phase equilibrium of Cu–Pb system under some specified pressure and temperature. On this basis, 60 min was selected as the final distillation time in this work which allows the system to approach the phase equilibrium sufficiently. In addition, it can also minimize the mass loss during experiment with increasing distillation time and reduce any errors associated the data that we obtained. The samples were collected from the volatile and residue, and then remelted, respectively, to determine the average components of the samples. The experimental data of the vapor–liquid phase equilibria for the Pb content in residuum (liquid phase) and volatiles (vapor phase) are listed in Table 2.



**Fig. 1.** Schematic diagram of the internal structure of the 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.

**Table 2**Experimental values of mole fraction  $x_{pb}$  and  $y_{pb}$  at temperature  $T$ , pressure  $P$  for the Cu–Pb alloy in vacuum distillation.<sup>a</sup>

Experiment condition			Content of Pb (mole fraction)	
Temperature (K)	Pressure (Pa)	Time (min)	$x_{pb}$	$y_{pb}$
1223	5	60	0.005367	0.999022
1273	5	60	0.002886	0.997066
1323	5	60	0.000834	0.993500
1373	5	60	0.000308	0.988018
1423	5	60	0.000015	0.981629

<sup>a</sup> Standard uncertainties  $u$  are  $u(T) = \pm 5\text{K}$ ,  $u(P) = 2\text{Pa}$ ,  $u(x) = \pm 0.000002$ , and  $u(y) = \pm 0.000002$ .

Considering the high temperatures and low vapor pressure during the experiment, the experimental VLE data we collected by this method will bring some certain deviations from ideal condition. They are mainly rising from the following aspects. The purity of the distillation products are both high ranging from 0.98 to 1, the deviations caused by the mass loss will be magnified in the final results. What's more, during the process of temperature rising to a certain degree, there will have be some mass metal evaporated from the liquid phase and the system vacuum degree will be outside the range of 10 ~ 15 Pa during this heating time. All the factors above are unavoidable so far.

### 3. Method

#### 3.1. VLE calculation

For the rare case of a low-pressure and ideal mixture system, Raoult's law applies to indicate the relationship of VLE which is expressed as [11,12]

$$x_i P_i^*(T) = y_i P \quad (1)$$

where  $x_i$  and  $y_i$  are the mole fraction of species  $i$  in the liquid phase and vapor phase, respectively;  $T$  is the temperature;  $P_i^*$  and  $P$  are the saturation pressure of species  $i$  and the pressure of the system, respectively, in terms of temperature.

Due to most of the liquid solutions for alloy systems cannot be considered as ideal, Raoult's law will give highly inaccurate results. For these systems under vacuum, the liquid phase is not an ideal solution but the pressure is low enough so that the vapor phase behaves as an ideal gas. In this case, the deviations are taken into account by incorporating a correction factor,  $\gamma$ , into Raoult's law to quantitatively consider the deviations from ideality. Therefore, Raoult's law can be modified as

$$x_i \gamma_i(x_i, T, P) P_i^*(T) = y_i P \quad (2)$$

where  $\gamma_i$  is activity coefficient of species  $i$  in terms of temperature, pressure and the mole fraction of species  $i$ .

For a binary alloy system  $i$ - $j$ ,

$$x_i + x_j = 1; y_i + y_j = 1 \quad (3)$$

$$P = x_i \gamma_i P_i^* + x_j \gamma_j P_j^* = x_i \gamma_i P_i^* + (1 - x_i) \gamma_j P_j^* \quad (4)$$

Download English Version:

<https://daneshyari.com/en/article/201522>

Download Persian Version:

<https://daneshyari.com/article/201522>

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