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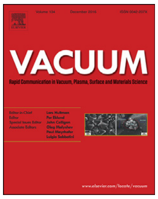

Experimental and modeling vapor-liquid equilibria: Separation of Bi from Sn by vacuum distillation

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ABSTRACT

The vapor-liquid equilibria (VLE) for Sn–Bi binary alloy at 5 Pa were carried out at distillation temperatures in the range of 1150 K–1550 K under vacuum condition. The experimental results indicate that Bi can be satisfactorily removed from crude Sn, the content of tin in liquid phase reaching more than 99.99 wt% at a temperature higher than 1300 K. The thermodynamic consistency of the experimental data was checked by Van Ness method. The Wilson equation is used to predicate VLE data. The results show good agreement with the experimental data, indicating that VLE phase diagram is feasible and practical for the process of vacuum distillation of alloys.

1. Introduction

The recycling of waste Sn-based alloys such as Sn–Sb, Sn–Bi, and Sn–Cu etc. is an important subject, not only for protection of the environment but also for recovery of valuable materials. Vacuum distillation, which has such advantages as high metal recovery, low impurities in recovering metal, good environmental protection and simple equipment, was widely used for separating and purifying crude metals [1].

Over past decades, much of experimental work has been done on the purification of crude materials by vacuum distillation, especially for Sn-based alloys, and good results were achieved [1–5]. VLE diagram is widely applied to predict and guide the operation of the distillation in chemical process [6,7]. In our previous work, we calculated the separation coefficient and composition of alloy systems to determine the degree of separation [8,9]. Although the calculation and application of vapor-liquid phase diagrams (*P*-*x* and *T*-*x* curves) of alloys in vacuum distillation were proposed [10,11], no experimental data on the vapor-liquid equilibrium are available for the Sn–Bi binary system till now. Little attention has been given to the application of phase diagrams for vapor-liquid equilibrium of alloys in vacuum distillation. VLE data are critical factors for designing and operating of separation processes for fluid mixtures. Therefore, it is of importance to experimentally investigate the phase equilibria of the Sn–Bi binary system in vacuum distillation.

The purpose of the present work is to investigate the vapor-liquid equilibrium for the Sn–Bi binary system in vacuum distillation. A consistency test of the calculated and experimental data were carried out using Herington test [12] and Van Ness method [13], respectively. Theoretical predications of VLE data were calculated using Wilson equation [14] and theory of VLE. The *T*-*x* phase diagram of Sn–Bi system was obtained which will bring an efficient and convenient way to analyze and predict the compositions of vapor and liquid phases for purification of Sn in vacuum distillation.

2. Experimental

2.1. Materials

The materials for the experiment from Yunnan tin Group Co., Ltd., China were used in this work. The Sn–Bi alloy was prepared

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using pure Sn (99.99 wt%) and Bi (99.99 wt%), with a content of Bi in the alloy of 0.2 mol fraction (30.56 wt%).

2.2. Apparatus and experimental procedure

A vertical vacuum furnace which made by Kunming Diboo Technology Co., Ltd. was used in this experiment. The schematic illustration of the furnace body is shown in Fig. 1. In the furnace, the crucible coil assembly was positioned the center of a water-cooled vacuum chamber which inner diameter 0.22 *m*, out diameter 0.27 *m* and 0.2 *m* depth. Distance between melt surface and condensing surface is 0.1 *m* as shown in Fig. 2. The maximum furnace temperature can be heated up to 1800 K. The absolute vacuum pressure reached less than 1 Pa before heating by operation of pump. Pressure in the vacuum chamber was evacuated to about 5 Pa by Rotary oil vacuum pump.

During the experiment, prepared Sn—Bi alloy melts of 80 g are made in the high-purity graphite crucible (99.998 wt % C) with an inner diameter 0.04 *m* and depth 0.04 *m* and are placed inside the internal diameter of 0.1 *m* crucible-coil. The vacuum pump started to work and vacuum furnace started to heat by induction heating, while the chamber is evacuated to 5 Pa. When complete melting, the chamber is heated to the preset temperature as soon as possible and then to keep the chamber at a constant temperature. Experimental investigation has been carried out in the temperature range between 1150 K and 1550 K. The saturated vapor pressure of Bi and Sn increases with temperature. The saturated vapor pressure follows the sequence of *Bi* > Sn at the same temperature. Therefore, Bi will exhibit a higher volatility than Sn. In the process of distillation, Bi evaporates from the surface of melt and condenses rapidly on the condenser, volatilization process as shown in Fig. 3.

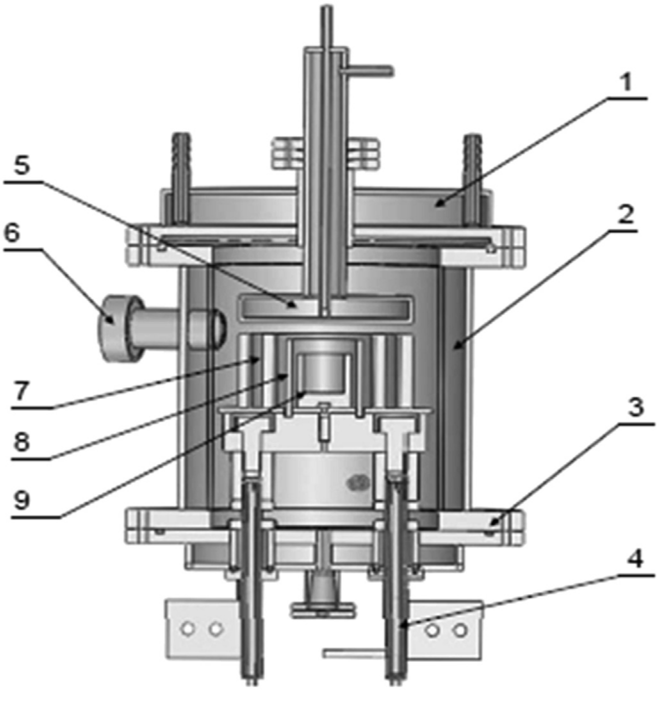

Fig. 1. Schematic diagram of the internal structure of the vertical vacuum furnace: 1 furnace lid; 2 furnace body; 3 furnace bottom; 4 electrode; 5 cold plate; 6 observation door; 7 heat holding cover; 8 heating unit; 9 graphite evaporator.

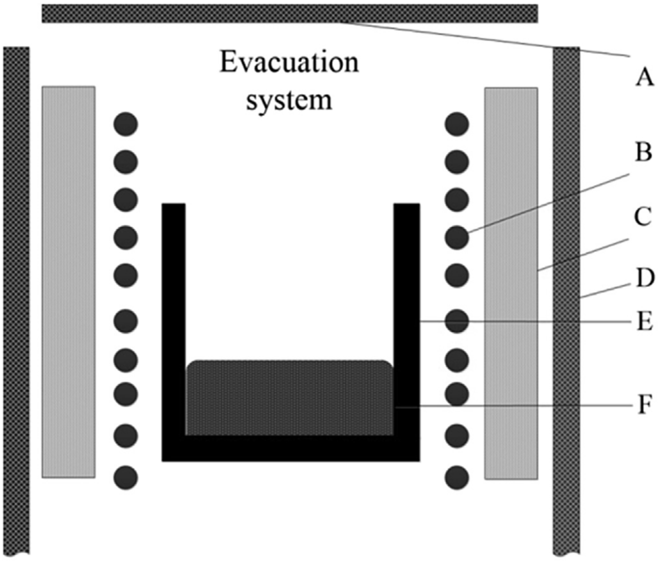

Fig. 2. Crucible arrangement: A – condensation tray; B – crucible-coil; C –heat holding cover; D – furnace body; E – graphite crucible; F – alloy melt.

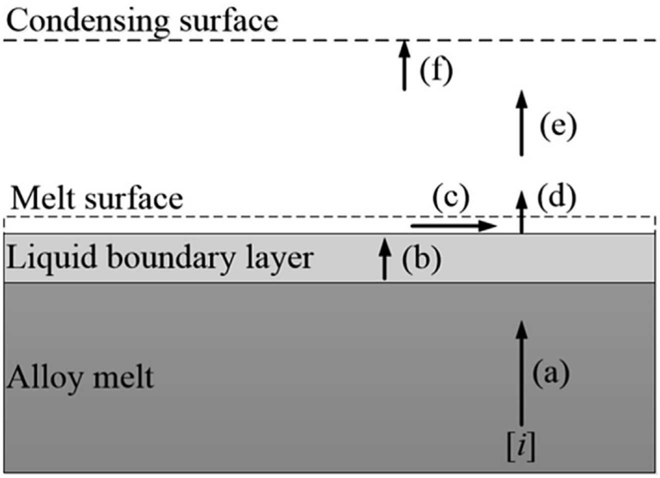

Fig. 3. Schematic of the mass transfer process of volatile impurity *i* from alloy melt:(a) Transport of an atom through the melt to the near metal boundary layer; (b) Transfer of the element through the metal boundary to the metal-gas interface; (c) Vaporization at the interface; (d) Transfer through a stagnant boundary layer on the gas side of the free metal surface; (e) Transport though the gas to the condensing site or the pumping gate; (f) Condensation or pumping out.

The vacuum metallurgical processes are, in principle, almost always made up of heterogeneous reactions. It is only very rarely that equilibrium exists within individual or between different phases, as only quasi-stationary states are attained. Equilibrium, therefore, should exist only at the phase boundary [15]. In order to calculate and promote the desired material transport which leads to the removal of impurities or to a separation of components via the gas or vapor phase, we hypothesized that the distillation processes will approach to a stable level with the extension of time. The distillation product approach to a stable level with the extension of time. It will be maintained long enough time to ensure the stability of system during experiment. The time increases gradually with decreasing distillation temperature. Finally, the distillation time was set from 360 min at 1150 K to 105 min at 1550 K. This condition will be maintained long enough time to ensure the stability of system. After completion of each distillation experiment, the system temperature was cooled to room temperature. The volatiles condensed on the stainless steel cold plate (connected to the circular cooling water) and the residues collected in the