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# Application of annexation principle of thermodynamic property of In-Pb-Sb and In-Bi-Pb melts①

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[Abstract] Based on the principle of annexation and phase diagrams, the structural units of In-Pb-Sb and In-Bi-Pb melts were determined, and their calculating models of mass action concentrations were formulated. Calculated results agree well with practical values, which in turn show that the models deduced can reflect the structural realities of corresponding melts and the principle of annexation is applicable not only to some binary metallic melts, but also to ternary metallic melts.

[Key words] principle of annexation; two phase melts; activity; mass action concentration
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## 1 INTRODUCTION

Alloys containing indium are used as electronic material. With rapid development of information technology, people pay much attention to indium containing alloys. Research on their thermodynamic properties also attracts great interest of metallurgist. Not only thermodynamic properties of each binary metallic melts included in ternary melts In-Pb-Sb and In-Bi-Pb have been systematically studied, but those of both ternary systems have also been comprehensively investigated[1~3]. These undoubtedly are reliable practical basis for the theoretical study. Since three binary metallic systems making up the two ternary melts are respectively binary melts involving eutectics (Pb-Sb, Bi-Pb), solid solution (In-Pb) and compound formation (In-Sb, In-Bi), i. e. three types of binary metallic system. How to formulate the calculating models of mass actions for these complicated melts is also an important theoretical problem. It is a coincidence that annexation of two kinds of solution in binary metallic melts has been investigated in references, which would surely be helpful to the study of the aforementioned two ternary melts. The aim of this paper is just to draw up the calculating models of mass action concentrations for these two ternary melts with help of the principle of annexation.

## 2 In-Pb-Sb MELTS

As this ternary system is composed of three binary systems Pb-Sb, In-Pb and In-Sb, so the formulation of calculating model is carried out with three binary systems.

### 2.1 Pb-Sb melts

According to the phase diagram[4], this binary system is a typical eutectic alloy. As its activities exhibit symmetrical negative deviation from Raoultian behavior, it is conjectured that in which a AB form metastable compound PbSb would appear. Hence, with regard to Pb-Sb melts, their structural units are Pb, Sb atoms as well as PbSb intermetallic compound (short-range order chemical cluster), and form two solutions Pb+ PbSb and Sb+ PbSb.

Putting the composition of the melts as $b = \sum x(\mathrm{Pb})$, $a = \sum x(\mathrm{Sb})$; the equilibrium mole fraction of every structural unit expressed by the composition of the melts as $x = x(\mathrm{Pb})$, $y = x(\mathrm{Sb})$, $z = x(\mathrm{PbSb})$; the mass action concentration of every structural unit as $N_1 = N(\mathrm{Pb})$, $N_2 = N(\mathrm{Sb})$, $N_3 = N(\mathrm{PbSb})$, then it gives the chemical equilibrium:

$$\mathrm{Pb(l)} + \mathrm{Sb(l)} = \mathrm{PbSb(l)}$$

$$K = N_3/(N_1N_2) \quad (1)$$

mass balance:

$$b = x + z \quad (2)$$

$$a = y + z \quad (3)$$

$$N_1 = x/b,\ N_2 = y/a,\ N_3 = KN_1N_2 \quad (4)$$

Substituting Eqn. (4) into Eqns. (2) and (3) gives

$$\left.\begin{aligned} N_1 + KN_1N_2/b &= 1 \\ N_2 + KN_1N_2/a &= 1 \end{aligned}\right\} \quad (5)$$

Summation of Eqn. (5) gives

$$K = ab(2 - N_1 - N_2)/[(a + b)N_1N_2] \quad (6)$$

Eqns. (5) and (6) are the calculating model of mass action concentrations for these melts, in which Eqn. (5) is used to calculate the mass action concentrations, while Eqn. (6) for evaluation of equilibrium constant on the condition of given measured activities ($N_1 = a(\mathrm{Pb})$, $N_2 = a(\mathrm{Sb})$).

According to the measured activities $a(\mathrm{Pb})$ and

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$a$(Sb) at 903, 929 and 1 073 K from Ref. [1] for Pb-Sb melts, and evaluating by Eqn. (6), it gives the equilibrium constants at the corresponding temperatures as $K_{903\,K}$ = 1.317 382, $K_{929\,K}$ = 1.284 266, $K_{1073\,K}$ = 1.246 13, and accordingly

$$\lg K = \frac{122.99}{T} - 0.019\,76$$

($r$ = 0.952 72)

$$\Delta G^{\ominus} = -2355.8 + 0.3785\,T \quad (J/mol) \qquad (7)$$

The calculated mass action concentrations are compared with measured activities as shown in Fig. 1.

Good agreement between calculated and measured values shows that the model is validly formulated.

## 2.2 In-Pb melts

According to the phase diagram[4], solid solution is formed in this binary system. Activities of these binary melts exhibit symmetrical positive deviation relative to Raoult's law. Then, it is conjectured that a AB form metastable compound InPb would form in these melts. Hence the structural units of these melts should be In, Pb atoms as well as InPb compound, and form two solutions In+ InPb and Pb+ InPb. As their calculating model of mass action concentrations are the same as Eqns. (1) ~ (6) for Pb-Sb melts. Using the measured activities $a$(In) and $a$(Pb) from Ref. [5, 6], the regressed relationships $K$-$T$ and $\Delta G^{\ominus}$ are given as

$$\lg K = \frac{-49.3316}{T} - 0.084\,312$$

(673~ 1 000 K)

$$\Delta G^{\ominus} = 944.93 + 1.615\,T \quad (J/mol) \qquad (8)$$

The comparison of calculated mass action concentrations and measured activities for these melts is shown in Fig. 2. Similarly, a good agreement between calculated and measured values testifies that the model is correctly deduced.

## 2.3 In-Sb melts

According to the phase diagram[4], there is one compound InSb with congruent melting point formed in these melts. It also forms eutectics both with In and Sb. Based on the coexistence theory of metallic melts[7], the structural units of these binary melts

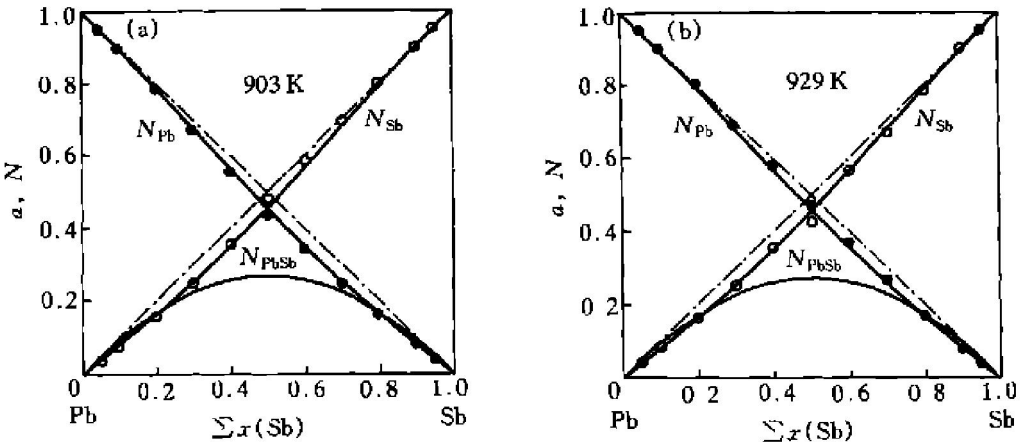


**Fig. 1** Comparison of calculated mass action concentrations with measured activities for Pb-Sb melts
(Solid line—calculated values; "●","○"—measured values; Pb—pure lead; Sb—pure antimony)

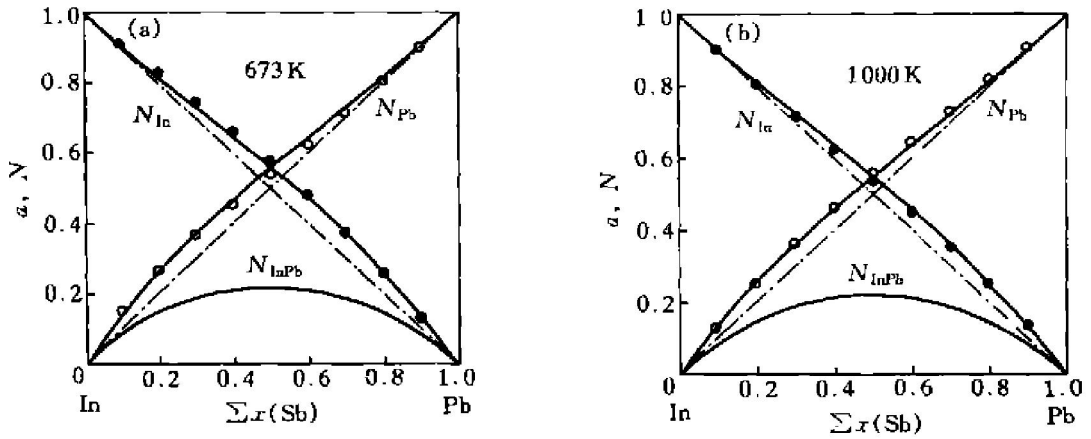


**Fig. 2** Comparison of calculated mass action concentrations (solid line) with measured activities("●","○") for In-Pb melts

are In, Sb atoms as well as InSb compound. Taking it as the fundamental to formulate the calculating model of mass action concentrations, the data of $N(\mathrm{In})$ and $N(\mathrm{Sb})$ should exhibit symmetrical negative deviation from Raoultian behavior. However, there isn't any symmetrical relationship between measured $a(\mathrm{In})$ and $a(\mathrm{Sb})$ from Ref. [8], which indicates that the structural units of these melts are inconsistent with the practice. Then, in consideration of the annexation principle for two kinds of solutions in binary metallic melts, the problem was going to be solved. i. e. in these melts, except InSb having congruent melting point (which promotes homogeneous solution to form), there is also an eutectic InSb+ In (two phase solutions) with a metastable compound $In_3Sb$ to form. From this, the structural units of these melts are In, Sb atoms as well as InSb and $In_3Sb$ compounds. Because the homogeneous solution involving compound formation is much stable than the two phase solution involving eutectic, so the resultant metallic melts should be still a homogeneous solution. Thus, assuming the composition of the melts as $a=\Sigma x(\mathrm{Sb})$, $b=\Sigma x(\mathrm{In})$; the mass action concentration of every structural unit after normalization as $N_1=N(\mathrm{In})$, $N_2=N(\mathrm{Sb})$, $N_3=N(\mathrm{InSb})$, $N_4=N(\mathrm{In_3Sb})$, $\Sigma x$ = sum of all equilibrium mole fractions, then it gives chemical equilibria:

$$\mathrm{In(l)+Sb(l)=InSb(l)}$$

$$K_1=\frac{N_3}{N_1N_2} \tag{9}$$

$$\mathrm{3In(l)+Sb(l)=In3Sb(l)}$$

$$K_2=\frac{N_4}{N_1^3N_2} \tag{10}$$

After making mass balance it gives

$$N_1+N_2+K_1N_1N_2+K_2N_1^3N_2=1 \tag{11}$$

$$aN_1-bN_2+(a-b)K_1N_1N_2+(3a-b)K_2N_1^3N_2=0 \tag{12}$$

$$1-(a+1)N_1-(1-b)N_2=K_1(a-b+1)N_1N_2+K_2(3a-b+1)N_1^3N_2 \tag{13}$$

Eqns. (11), (12) and (13) mentioned above are the calculating model of mass actions for these melts (i. e. the normal calculating model without interference with other element), in which Eqns. (11) and (12) are used to calculate the mass action concentrations, while Eqn. (13) used to regress equilibrium constants under the condition of given measured activities ($a(\mathrm{In})=N_1$, $a(\mathrm{Sb})=N_2$). Using measured activities from Ref. [1], inserting them into Eqn. (13) and after regressing it gives equilibrium constants and $\Delta G^{\ominus}$ at 1 073 K respectively as $K_{\mathrm{InSb}}=3.218\,15$ ($\Delta G^{\ominus}=-10\,432.78$ J/mol), $K_{\mathrm{In_3Sb}}=3.470\,026$ ($\Delta G^{\ominus}=-11\,105.4$ J/mol), ($F=1\,079.656$; $R=0.995\,858$).

Substituting the thermodynamic data into Eqns. (11) and (12), after computing, it also gives good agreement between calculated and measured values, showing that the structural units determined and the calculating model formulated could represent the structural reality of the melts. It also testifies that in these melts actually occurred the annexation process of the two phase melts involving eutectic by the homogeneous melts involving compound formation with the resultant formation of $In_3Sb$. Due to limited space, here the figure of comparison of calculated and measured values was omitted.

However, as both Pb-Sb and In-Pb are two phase solutions, their effect is much greater than that of the homogeneous In-Sb melts. Under conditions of forming In-Pb-Sb ternary melts, In-Sb homogeneous melts should also be annexed by the former two solutions (two phase) and changed into a two phase solution. Since the reaction mechanism of two phase solution is different from a homogeneous one, so its thermodynamic parameters should also be evaluated by the following calculating model of two phase solutions (In+ InSb+ $In_3Sb$ and Sb+ InSb+ $In_3Sb$) (all symbols are the same as the calculating model of homogeneous solutions):

$$b=x+z_1+3z_2,\quad a=y+z_1+z_2, \tag{14}$$

$$N_1=\frac{y}{b},\quad N_2=\frac{y}{a}, \tag{15}$$

$$z_1=K_1N_1N_2,\quad z_2=K_2N_1^3N_2 \tag{16}$$

$$N_1+(K_1N_1N_2+3K_2N_1^3N_2)/b=1,$$

$$N_2+(K_1N_1N_2+K_2N_1^3N_2)/a=1 \tag{17}$$

$$ab(2-N_1-N_2)=K_1(a+b)N_1N_2+K_2(3a+b)N_1^3N_2 \tag{18}$$

Eqns. (17) and (18) are the calculating models of mass action concentrations for In-Sb melts suitable for the application to the condition of two phase melts, in which Eqn. (17) is used to calculate the mass action concentrations, while Eqn. (18) used to regress equilibrium constants. Using the measured activities at 1 073 K from Ref. [1], the equilibrium constants and $\Delta G^{\ominus}$ are respectively regressed as $K_{\mathrm{InSb}}=3.328\,04$ ($\Delta G^{\ominus}=-10\,732.5$ J/mol); $K_{\mathrm{In_3Sb}}=0.849\,118$ ($\Delta G^{\ominus}=1\,459.9$ J/mol).

The calculated results were also agreed well with practice, here they are omitted.

### 2.4 In-Pb-Sb ternary melts

From the three binary melts preceding discussed, it is known that these melts are inhomogeneous melts. Their structural units are In, Pb and Sb atoms as well as four compounds InPb, InSb, $In_3Sb$ and PbSb, and they form three solutions In+ InPb+ InSb+ $In_3Sb$, Pb+ InPb+ PbSb and Sb+ InSb+ $In_3Sb$+ PbSb.

Assuming the composition of the melts as $a=\Sigma x(\mathrm{In})$, $b=\Sigma x(\mathrm{Pb})$, $c=\Sigma x(\mathrm{Sb})$, the equilibrium mole fraction of every structural unit expressed by the

composition of the melts as $x = x(\mathrm{In})$, $y = x(\mathrm{Pb})$, $z = x(\mathrm{Sb})$, $u_1 = x(\mathrm{InPb})$, $u_2 = x(\mathrm{InSb})$, $u_3 = x(\mathrm{In_3Sb})$, $u_4 = x(\mathrm{PbSb})$; the mass action concentration of every structural unit as $N_1 = N(\mathrm{In})$, $N_2 = N(\mathrm{Pb})$, $N_3 = N(\mathrm{Sb})$, $N_4 = N(\mathrm{InPb})$, $N_5 = N(\mathrm{InSb})$, $N_6 = N(\mathrm{In_3Sb})$, $N_7 = N(\mathrm{PbSb})$, then it gives chemical equilibria:

$$\mathrm{In(l)} + \mathrm{Pb(l)} = \mathrm{InPb(l)} \qquad K_1 = \frac{N_4}{N_1 N_2}$$

$$\Delta G^{\ominus} = 944.93 + 1.615\,T \quad (\mathrm{J/mol}) \qquad (19)$$

$$\mathrm{In(l)} + \mathrm{Sb(l)} = \mathrm{InSb(l)} \qquad K_2 = \frac{N_5}{N_1 N_3}$$

$$\Delta G^{\ominus} = -10732.5 \quad (\mathrm{J/mol}) \quad (1073\,\mathrm{K}) \qquad (20)$$

$$3\mathrm{In(l)} + \mathrm{Sb(l)} = \mathrm{In_3Sb(l)} \qquad K_3 = \frac{N_6}{N_1^3 N_3}$$

$$\Delta G^{\ominus} = 1459.9 \quad (\mathrm{J/mol}) \quad (1073\,\mathrm{K}) \qquad (21)$$

$$\mathrm{Pb(l)} + \mathrm{Sb(l)} = \mathrm{PbSb(l)} \qquad K_4 = \frac{N_7}{N_2 N_3}$$

$$\Delta G^{\ominus} = -2355.8 + 0.3785\,T \quad (\mathrm{J/mol}) \qquad (22)$$

mass balance:

$$a = x + u_1 + u_2 + 3u_3,$$
$$b = y + u_1 + u_4,$$
$$c = z + u_2 + u_3 + u_4 \qquad (23)$$

$$N_1 + (K_1 N_1 N_2 + K_2 N_1 N_3 + 3K_3 N_1^3 N_3)/a = 1 \qquad (24)$$

$$N_2 + (K_1 N_1 N_2 + K_4 N_2 N_3)/b = 1 \qquad (25)$$

$$N_3 + (K_2 N_1 N_3 + K_3 N_1^3 N_3 + K_4 N_2 N_3)/c = 1 \qquad (26)$$

Summation of Eqns. (24) ~ (26) gives

$$abc(3 - N_1 - N_2 - N_3) = c(a+b)K_1 N_1 N_2 + b(c+a)K_2 N_1 N_3 + b(3c+a)K_3 N_1^3 N_3 + a(b+c)K_4 N_2 N_3 \qquad (27)$$

Eqns. (24), (25), (26) and (27) are the calculating model of mass action concentrations for ternary melts In-Pb-Sb, in which Eqns. (24), (25) and (26) are used to calculate the mass action concentrations, while Eqn. (27) used to regress the equilibrium constants.

The calculated mass action concentrations at 1073 K for these ternary melts are compared with measured activities from Ref. [1] as shown in Fig. 3. Fig. 3 shows that the agreement between them is quite good, and the calculating model formulated could embody the structural reality of these melts, meanwhile, annexation principle is applicable to the ternary melts.

## 3 In-Bi-Pb MELTS

Similarly, as the ternary system composed of three binary systems Bi-Pb, In-Pb and Bi-In, the formulation of calculating model starts with three binary systems.

### 3.1 Bi-Pb melts

According to the phase diagram[4], in the binary

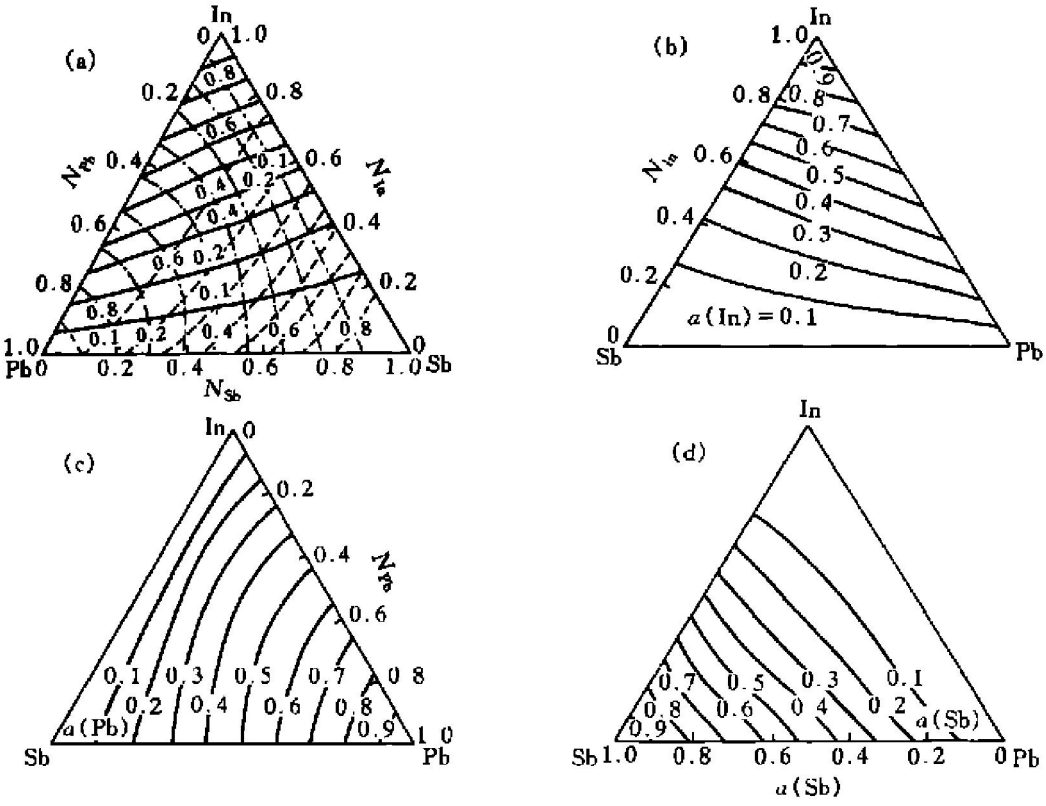


**Fig. 3** Comparison of calculated mass action concentrations $N(\mathrm{In})$, $N(\mathrm{Pb})$ and $N(\mathrm{Sb})$ (a) with measured activities $a(\mathrm{In})$ (b), $a(\mathrm{Pb})$ (c) and $a(\mathrm{Sb})$ (d) for In-Pb Sb melts at 1073 K

system an eutectic alloy is formed. Its activities exhibit symmetrical negative deviation from Raoultian behavior, hence the preceding calculating model of two phase Pb-Sb melts, Eqns. (5) and (6), are also applicable to this case, for this reason, separate deduction is not needed. According to Ref. [9], the Gibbs free energy for the chemical reaction of BiPb compound formation is

$$Bi(l) + Pb(l) = BiPb(l)$$

$$\Delta G^{\ominus} = -2937.63 - 1.198T \quad (J/mol) \quad (28)$$

Using Eqn. (5) and thermodynamic parameter of Eqn. (28), the calculated mass action concentrations are compared with the measured activities at 1 223 K as shown in Fig. 4. Good agreement between calculated and measured values shows that the calculating model could reflect the structural reality of these melts.

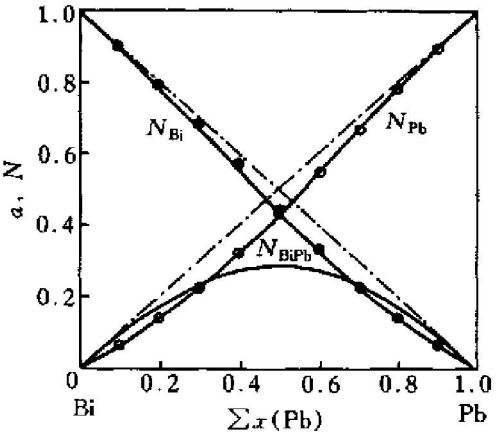


**Fig. 4** Comparison of calculated mass action concentrations (solid line) with measured activities ("•", "○") for Bi-Pb melts at 1 223 K

### 3.2 Bi-In melts

According to the phase diagram[4], there are three compounds BiIn, $Bi_5In_3$ and $BiIn_2$ formed in the system, in which BiIn and $BiIn_2$ have congruent melting points, while $Bi_5In_3$ is a peritectic. Hence, when these melts are a homogeneous solution, their structural units are Bi, In atoms as well as BiIn, $Bi_5In_3$ and $BiIn_2$ compounds.

However, similarly as In-Pb-Sb melts, both Bi-Pb and In-Pb systems are two phase melts in In-Bi-Pb melts, the effect of these two melts is greater than that of the homogeneous Bi-In melts. Hence in case of forming a ternary In-Bi-Pb melts, Bi-In homogeneous melts will also be annexed by the two solutions mentioned above (two phase) and changed to two phase solutions. After repeatedly examination, it was found that in case of two phase solutions, peritectic $Bi_5In_3$ will not form again. So the structural units of these melts are Bi, In atoms as well as BiIn and $BiIn_2$ compounds. Putting the composition of the melts as $a = \Sigma x(Pb)$, $b = \Sigma x(In)$; the mass action concentration of every structural unit as $N_1 = N(Bi)$, $N_2 = N(In)$, $N_3 = N(BiIn)$, $N_4 = N(BiIn_2)$, then it gives chemical equilibria:

$$Bi(l) + In(l) = BiIn(l) \qquad K_1 = \frac{N_3}{N_1 N_2} \quad (29)$$

$$Bi(l) + 2In(l) = BiIn_2(l) \qquad K_2 = \frac{N_4}{N_1 N_2^2} \quad (30)$$

After making mass balance it gives the calculating model of mass action concentrations for two phase melts as

$$N_1 + (K_1 N_1 N_2 + K_2 N_1 N_2^2)/b = 1 \quad (31)$$

$$N_2 + (K_1 N_1 N_2 + 2K_2 N_1 N_2^2)/a = 1 \quad (32)$$

$$ab(2 - N_2 - N_2) = (a + b)K_1 N_1 N_2 + (a + 2b)K_2 N_1 N_2^2 \quad (33)$$

Eqns. (31), (32) and (33) are the calculating model of mass action concentrations for two phase melts, in which Eqns. (31) and (32) are used for the calculation of the mass action concentrations, while Eqn. (33) used for regression of equilibrium constants in case of given measured activities ($N_1 = a(Bi)$, $N_2 = a(In)$).

By the use of Eqn. (33) and the measured activities from Ref. [10], the regressed relationships $K$-$T$ and $\Delta G^{\ominus}$ at 900~ 1 200 K are given respectively as

$$\lg K_{BiIn} = \frac{363.99}{T},$$

$$\Delta G^{\ominus} = -6972.069 + 1.656T \quad (J/mol)$$

(900~ 1 200 K) (34)

$$\lg K_{BiIn_2} = \frac{-99.85}{T}$$

$$\Delta G^{\ominus} = 1912.584 + 8.84T \quad (J/mol)$$

(900~ 1 200 K) (35)

The comparison of calculated mass action concentrations with measured activities at 900 K and 1 200 K is given respectively in Fig. 5. As seen Fig. 1, the calculated results agree well with the practice, showing that the calculating model for two phase Bi-In melts could embody the structural characteristics of the melts after annexation.

### 3.3 In-Bi-Pb ternary melts

From the above discussions, it is clear that In-Bi-Pb system are inhomogeneous melts, their structural units are In, Bi and Pb atoms as well as four compounds BiIn, $BiIn_2$, InPb and BiPb, and they form three solutions In+ BiIn+ $BiIn_2$+ InPb, Bi+ BiIn+ $BiIn_2$+ BiPb and Pb+ InPb+ BiPb.

Assuming the composition of the melts as $a = \Sigma x(In)$, $b = \Sigma x(Bi)$, $c = \Sigma x(Pb)$; the equilibrium mole fraction of every structural unit expressed by the composition of the melts as $x = x(In)$, $y = x(Bi)$, $z = x(Pb)$, $u_1 = x(BiIn)$, $u_2 = x(BiIn_2)$, $u_3 = x(InPb)$, $u_4 = x(BiPb)$; the mass action concentration of every structural unit as $N_1 = N(In)$, $N_2 = N(Bi)$, $N_3 = N(Pb)$, $N_4 = N(BiIn)$, $N_5 = N(Bi$-

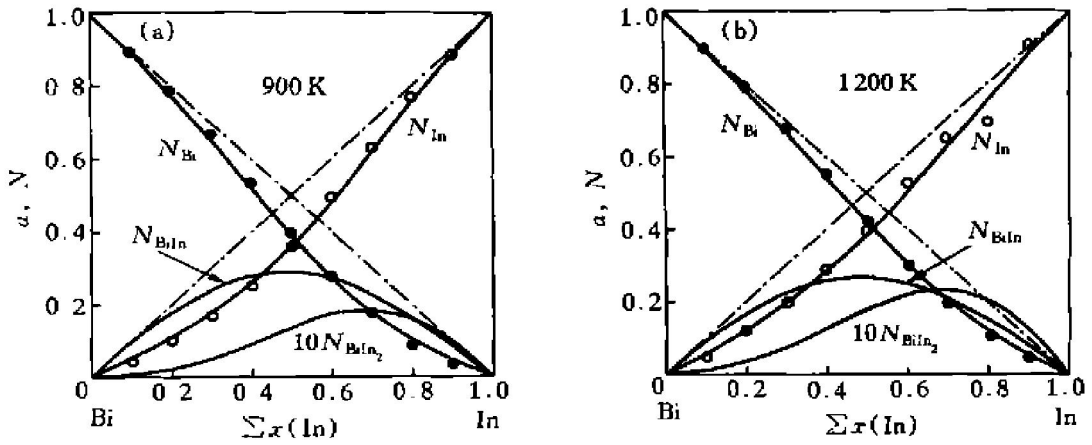


**Fig. 5** Comparison of calculated mass action concentrations (solid line) with measured activities ("●","○") for Bi-In melts

$In_2$), $N_6 = N(InPb)$, $N_7 = N(BiPb)$, then it gives chemical equilibria:

$$Bi(l) + In(l) = BiIn(l) \qquad K_1 = \frac{N_4}{N_1 N_2}$$

$$\Delta G^{\ominus} = -6972.069 + 1.656T \quad (J/mol) \quad (36)$$

$$Bi(l) + 2In(l) = BiIn_2(l) \qquad K_2 = \frac{N_5}{N_1^2 N_2}$$

$$\Delta G^{\ominus} = 1912.584 + 8.84T \quad (J/mol) \quad (37)$$

$$In(l) + Pb(l) = InPb(l) \qquad K_3 = \frac{N_6}{N_1 N_3}$$

$$\Delta G^{\ominus} = 944.93 + 1.615T \quad (J/mol) \quad (38)$$

$$Bi(l) + Pb(l) = BiPb(l) \qquad K_4 = \frac{N_7}{N_2 N_3}$$

$$\Delta G^{\ominus} = -2937.63 - 1.198T \quad (J/mol) \quad (39)$$

mass balance:

$$a = x + u_1 + 2u_2 + u_3,$$
$$b = y + u_1 + u_2 + u_4,$$
$$c = z + u_3 + u_4 \quad (40)$$

$$N_1 + (K_1 N_1 N_2 + 2K_2 N_1^2 N_2 + K_3 N_1 N_3)/a = 1 \quad (41)$$

$$N_2 + (K_1 N_1 N_2 + K_2 N_1^2 N_2 + K_4 N_2 N_3)/b = 1 \quad (42)$$

$$N_3 + (K_3 N_1 N_3 + K_4 N_2 N_3)/c = 1 \quad (43)$$

Summation of Eqns. (39) ~ (41) gives

$$(abc)(3 - N_1 - N_2 - N_3) = c(a+b)K_1 N_1 N_2 + c(a+2a)K_2 N_1^2 N_2 + b(c+a)K_3 N_1 N_3 + a(b+c)K_4 N_2 N_3 \quad (44)$$

The above mentioned Eqns. (39), (40), (41) and (42) are the calculating models of mass action concentrations for ternary melts In-Bi-Pb, in which Eqns. (39), (40) and (41) are used for calculation of mass action concentrations, while Eqn. (42) used for regression of equilibrium constants.

The calculated mass action concentrations for these ternary melts at 773 K are compared with the measured activities from Ref. [2] in Fig. 6. It is seen from the figure that agreement between calculated and measured values is quite well, showing that the model formulated could reflect the structural reality of these melts, and in the meantime testifying that the annexation principle is also applicable to these ternary melts.

From the above two examples, it is evident that a pair of two phase melts In-Pb and Pb-Sb can annex homogeneous melts In-Sb, making the latter change to a two phase solution and merge into an inhomogeneous ternary melts In-Pb-Sb. Similarly a pair of two phase melts In-Pb and Bi-Pb annex a homogeneous melts Bi-In, making the latter change to a two phase solution and also merge into an inhomogeneous ternary melts In-Bi-Pb. Thus, a pair of two phase melts could annex a homogeneous melts, it is the common principle which determined the structural units of the above mentioned two ternary melts.

## 4 CONCLUSIONS

1) A pair of two phase melts could annex the homogeneous melts. Annexation is an important principle for determination of the structural units for some binary melts, and also is indispensable to ascertaining the structural units of some ternary melts.

2) According to the annexation principle and phase diagrams, the structural units of two ternary melts In-Pb-Sb and In-Bi-Pb have been ascertained, and the calculating models of mass action concentrations for these two melts have been formulated. The results of calculation agree well with the practice, showing that the models formulated could embody the structural reality of both melts.

3) The reaction mechanism of two phase melts is different from a homogeneous one, hence their thermodynamic parameters should be determined separately.

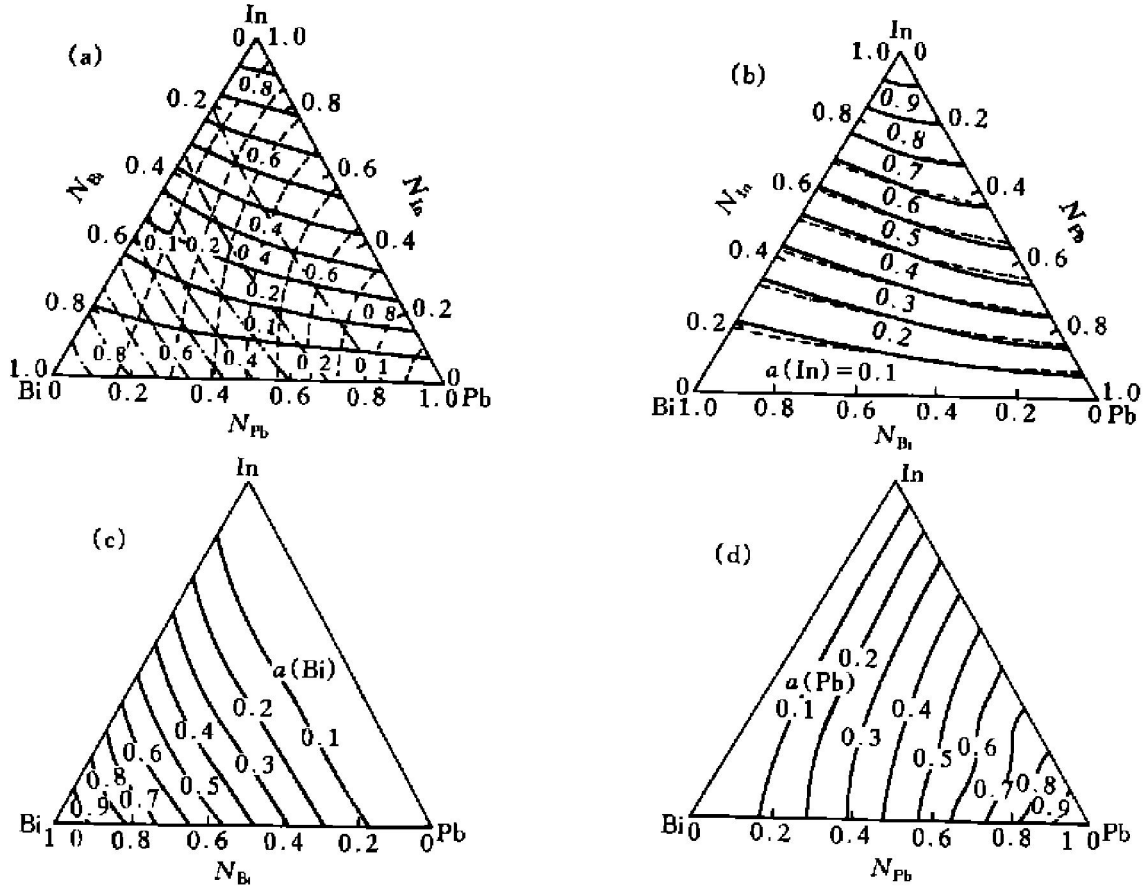


**Fig. 6** Comparison of calculated mass action concentrations $N$(In), $N$(Bi) and $N$(Pb) (a) with measured activities $a$(In) (b), $a$(Bi) (c) and $a$(Pb) (d) for In-Bi melts at 773 K

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