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# Calculation Models of Mass Action Concentrations for Metallic Melts Involving Monotectic

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**Abstract :** Based on the phase diagrams, measured activities as well as$\Delta G^{m}$ and $\Delta G^{xs}$, calculating models of mass action concentrations for metallic melts involving monotectic have been formulated. The calculated results agree with practice on the whole, showing that the models deduced generally can reflect the structural characteristics of these melts. The metastable compounds formed in the melts are of the types $A_2B_3$, $AB_2$, $A_2B_3$or AB and $A_2B_3$+AB etc..

**Key words:** metallic melts; monotectic; activity; mass action concentration

## 1 Introduction

In preceding paper [1] binary metallic melts have been classified according to their phase diagrams as six types: metallic melts involving compound formation, involving peritectic, involving saturated phase, involving solid solution, involving eutectic and involving continuous solid solution. Binary metallic melts involving monotectic have been put into binary metallic melts involving eutectic. From the current point of view, it is not correct, as binary metallic melts involving monotectic have their own characteristics. It is seen from **table 1** that the differences of atomic mass of two elements constituting this type of melts are all considerably great. Hence the effect of relative density caused by the difference of atomic mass seems to be important factor leading to the formation of this type of melts. Originally, binary metallic melts involving eutectic and monotectic is about 9% of all binary metallic systems. At present, it is known that the binary metallic systems involving monotectic is 2.7%, so binary metallic systems involving eutectic should be 6.3 % of all binary metallic systems. This type of binary metallic melts over a wide range of compositions on account of gravity force obviously separates into two solutions [2], this undoubtedly will impose restriction on chemical reactions between them due to the resistance of diffusion across the boundary layer. According to the knowledge of structural chemistry [3], in natural world, only inert gases He, Ne, Ar, Kr, Xe etc., can exist in form of atoms; At standard state, all other elements exist in form of complex of atoms, molecule or crystal. Judging from this, in spite of separating into two solutions, chemical reactions between two elements in these melts are still inevitable. For this reason, the goal of this paper is to study chemical reactions and their thermodynamic properties in melts involving monotectic which separate into two solutions.

**Table 1 Binary alloys involving monotectic and relative atomic mass of their components**

| Binary alloy | Atomic mass | | Binary alloy | Atomic mass | | Binary alloy | Atomic mass | |
|---|---|---|---|---|---|---|---|---|
| A–B | A | B | A–B | A | B | A–B | A | B |
| Li–Na | 6.94 | 22.99 | Ti–W | 47.9 | 183.8 | Zn–Bi | 65.3 | 208.98 |
| Na–Ca | 22.99 | 40.08 | V–Cu | 50.941 | 63.54 | Zn–Tl | 65.3 | 204.3 |
| Al–Cd | 26.96 | 112.4 | V–In | 50.941 | 114.82 | Zn–Pb | 65.3 | 207.2 |
| Al–In | 26.96 | 114.82 | Cr–Lu | 51.996 | 174.97 | Ga–Cd | 69.72 | 112.4 |
| Al–Pb | 26.96 | 207.2 | Cr–Sn | 51.996 | 118.6 | Ga–Tl | 69.72 | 204.3 |
| Al–Bi | 26.96 | 208.98 | Ni–Ag | 58.7 | 107.87 | Ga–Pb | 69.72 | 207.2 |
| Ti–Ce | 47.9 | 140.12 | Ni–Pb | 58.7 | 207.2 | Ga–Bi | 69.72 | 208.98 |
| Ti–Cd | 47.9 | 112.4 | Cu–Hg | 63.54 | 200.5 | Nb–U | 92.91 | 238.03 |
| Ti–Nd | 47.9 | 144.2 | Cu–Tl | 63.54 | 204.3 | | | |
| Ti–Y | 47.9 | 88.91 | Cu–Pb | 63.54 | 207.2 | | | |

## 2 Cu–Pb melts

It is seen from the activities $a_{Cu}$ and $a_{Pb}$ as well as thermodynamic parameters $\Delta G^m$ and $\Delta G^{xs}$ [4] in **table 2** that the activities of Cu–Pb melts exhibit positive deviation relative to Raoult's law, and at $\Sigma x_{Pb}=0.6$, $\Delta G^m$ has minimum value , which shows that these melts are two phase solution indeed. Hence according to the consistency of thermodynamic properties with the structure of binary metallic melts [1], it is conjectured that there should be a $A_2B_3$ form short-range chemical cluster formation in these melts, i.e., $Cu_2Pb_3$. Consequently, these melts consist of Cu, Pb atoms and $Cu_2Pb_3$ compound, and form two solutions $Cu+Cu_2Pb_3$ and $Pb+Cu_2Pb_3$. Thus putting the composition of the melts as $a=\Sigma x_{Pb}$, $b=\Sigma x_{Cu}$, the equilibrium mole fraction of every structural unit expressed by the composition of the melts as $x=x_{Cu}$, $y=x_{Pb}$, $z=x_{CuPb_3}$, and the mass action concentration of every structural unit as $N_1=N_{Cu}$, $N_2=N_{Pb}$, $N_3=N_{CuPb_3}$, then it gives chemical equilibrium:

$$2Cu_{(l)}+3Pb_{(l)}=Cu_2Pb_{3(l)} \qquad K=\frac{N_3}{N_1^2N_2^3} \tag{1}$$

Mass balance:

$$b=x+2z \tag{2}$$

$$a=y+3z \tag{3}$$

$$N_1=x/b, N_2=y/a, N_3=KN_1^2N_2^3 \tag{4}$$

Substituting equation (4) into equations (2) and (3) gives:

$$N_1+2KN_1^2N_2^3/b=1, N_2+3KN_1^2n_2^3/a=1 \tag{5}$$

Summation of equation (5) gives:

$$N_1+N_2+KN_1^2N_2^3\left(\frac{3}{a}+\frac{2}{b}\right)=2 \tag{6}$$

$$K=ab(2-N_1-N_2)/[(2a+3b)N_1^2N_2^3] \tag{7}$$

Equations (5) and (7) are the calculating model of mass action concentrations for these melts, in which equation (5) is used to calculate the mass action concentrations, while equation (7) to evaluate equilibrium constant on condition of given measured activities ($N_1=a_{Cu}$, $N_2=a_{Pb}$). Using measured activities at 1 473 K in table 2 [4], the equilibrium constant has been evaluated as $K=0.184\,948$, Substituting it into equation (5), the results of calculation are given in **figure 1**. It can be seen from the figure that the agreement between calculated and measured values is satisfactory, showing that the model formulated can reflect the structural reality of these melts. In the meantime, it is seen that the model deduced is completely identical with that of binary metallic melts involving eutectic containing $A_2B_3$ form short-range order chemical cluster. But the agreement between calculated and measured values is not as good as that of the latter, which is possibly arisen from the macroscopic separation of these melts into two distinct solutions and restriction of chemical reactions due to resistance of diffusion across the boundary layer, as a result of this, chemical reactions are not easy to completely reach equilibrium; while melts involving eutectic, in spite of that they are also a two phase solution, but they don't separate distinctly into two layers. In other words, they are still mechanically mixed together, hence chemical reactions between two components can easily reach equilibrium.

**Table 2 Measured activities as well as $\Delta G^m$ and $\Delta G^{xs}$ of Cu-Pb melts at 1 473 K**

| $\Sigma x_{Pb}$ | $a_{Cu}$ | $a_{Pb}$ | $\Delta G^m$/(J·mol$^{-1}$) | $\Delta G^{xs}$/(J·mol$^{-1}$) |
|---|---|---|---|---|
| 0.1 | 0.912 | 0.417 | −2 093.4 | 1 892.4 |
| 0.2 | 0.852 | 0.630 | −2 700.5 | 3 433.2 |
| 0.3 | 0.832 | 0.683 | −2 981.0 | 4 505.0 |
| 0.4 | 0.815 | 0.709 | −3 190.3 | 5 057.7 |
| 0.5 | 0.791 | 0.736 | −3 320.1 | 5 170.7 |
| 0.6 | 0.755 | 0.764 | −3 362.0 | 4 886.0 |
| 0.7 | 0.691 | 0.800 | −3 274.1 | 4 211.9 |
| 0.8 | 0.577 | 0.848 | −2 960.1 | 3 169.4 |
| 0.9 | 0.370 | 0.913 | −2 219.0 | 1 762.6 |

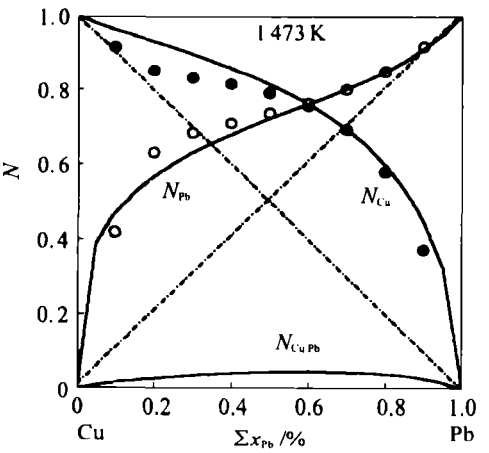


**Figure 1 Comparison of calculated mass action concentrations(—) with measured activities (•○) for Cu-Pb melts at 1 473 K.**

## 3 Al–In, Cu–Tl and Zn–Bi melts

**Table 3** is a list of $\Delta G^m$ and $\Delta G^{xs}$ [4] for three binary metallic melts Al–In, Cu–Tl and Zn–Bi. It is seen from the table that at $\Sigma x_{In-Bi}=0.6$—0.7, $\Delta G^m$ exhibits minimum values. Similarly, according to the consistency of thermodynamic properties with the structure of binary metallic melts [1], it is conjectured that in these melts $AB_2$ form compound would possibly formed, i. e., $AlIn_2$, $CuTl_2$ and $ZnBi_2$. Taking Al–In melts as an example, the structural units of which should be Al, In atoms

Table 3 $\Delta G^m$ and $\Delta G^{xs}$ of Al–In, Cu–Tl and Zn–Bi melts J·mol$^{-1}$

| $\Sigma x_{In-Bi}$ | Al–In(1 173 K) | | Cu–Tl(1 573 K) | | Zn–Bi(873 K) | |
|---|---|---|---|---|---|---|
| | $\Delta G^m$ | $\Delta G^{xs}$ | $\Delta G^m$ | $\Delta G^{xs}$ | $\Delta G^m$ | $\Delta G^{xs}$ |
| 0.1 | −1 193.24 | 1 976.17 | −1 951.05 | 2 302.74 | −674.07 | 1 687.28 |
| 0.2 | −1 157.01 | 3 424.80 | −2 570.70 | 3 977.46 | −1 130.44 | 2 503.71 |
| 0.3 | −1 574.24 | 4 387.77 | −2 939.13 | 5 053.47 | −1 595.17 | 2 838.65 |
| 0.4 | −1 662.16 | 4 906.93 | −3 248.96 | 5 560.07 | −2 022.22 | 2 863.77 |
| 0.5 | −1 745.90 | 5 015.79 | −3 537.85 | 5 534.95 | −2 352.98 | 2 679.55 |
| 0.6 | −1 818.07 | 4 752.02 | −3 763.93 | 5 045.09 | −2 545.57 | 2 344.61 |
| 0.7 | −1 842.19 | 4 119.81 | −3 830.92 | 4 165.87 | −2 545.57 | 1 892.43 |
| 0.8 | −1 758.46 | 3 123.35 | −3 575.53 | 2 976.81 | −2 285.99 | 1 348.15 |
| 0.9 | −1 427.70 | 1 745.90 | −2 696.30 | 1 557.49 | −1 649.60 | 711.76 |

as well as $AlIn_2$ compound (or short-range chemical cluster), and form two solutions Al+$AlIn_2$ and In+$AlIn_2$. From this, putting the composition of the melts as $a=\Sigma x_{In}$, $b=\Sigma x_{Al}$, the equilibrium mole fraction of every structural unit expressed by the composition of the melts as $x=x_{Al}$, $y=x_{In}$, $z=x_{AlIn_2}$, and the mass action concentration of every structural unit as $N_1=N_{Al}$, $N_2=N_{In}$, $N_3=N_{AlIn_2}$, then it gives chemical equilibrium:

$$Al_{(l)} + 2In_{(l)} = AlIn_{2(l)} \qquad K=\frac{N_3}{N_1N_2^2} \tag{8}$$

Mass balance:

$$b=x+z \tag{9}$$

$$a=y+2z \tag{10}$$

$$N_1=x/b,\quad N_2=y/a,\quad N_3=KN_1N_2^2 \tag{11}$$

Substituting equation (11) into equations (9) and (10) gives:

$$N_1+KN_1N_2^2/b=1,\quad N_2+2KN_1N_2^2/a=1 \tag{12}$$

Summation of equation (12) gives:

$$N_1+N_2+KN_1N_2^2\left(\frac{2}{a}+\frac{1}{b}\right)=2 \tag{13}$$

$$K=ab(2-N_1-N_2)/[(a+2b)N_1N_2^2] \tag{14}$$

Equations (12) and (14) are the calculating model of mass action concentrations for these melts, in which equation (12) is used to calculate the mass action concentrations, while equation (14) for evaluation of equilibrium constant on condition of given measured activities ($N_1=a_{Al}$, $N_2=a_{In}$).

Substituting the measured activities of Al–In, Cu–Tl and Zn–Bi melts from reference [4] into equation (14) gives their corresponding equilibrium constants for compound formation as $K_{AlIn}$(1 173 K)=0.088 734 6, $K_{CuTl}$(1 573 K)=0.191 005 and $K_{ZnBi}$(873 K)=0.289 377. Using these equilibrium constants, the calculated mass action concentrations are compared with the measured activities as shown in **figure 2**. It is seen from three figures that the agreement between calculated and measured values on the whole is satisfactory, showing that the model deduced can reflect the structural reality of the above mentioned three melts. However, there are also some points in the figures, which do not confirm enough with measured values. As mentioned before, their presence is possible due to distinct separation of the melts into two solutions, as a result of this, chemical reactions between two components are not easily to reach equilibrium.

## 4 Al–Zn melts

There are different compounds formed in Al–Zn

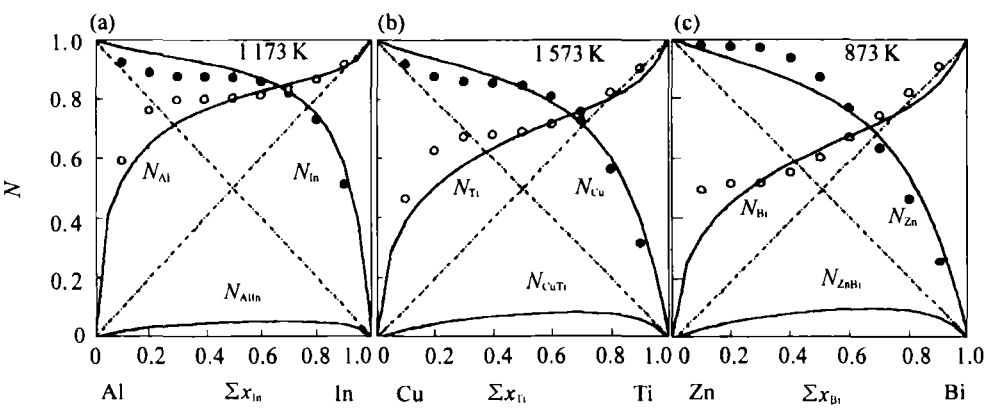


**Figure 2 Comparison of calculated mass action concentrations(—) with measured activities (•○), (a) Al–In melts(1 173 K); (b) Cu–Tl melts(1 573 K); (c) Zn–Bi melts (873 K).**

melts at different temperatures. For example, at 653 K the compound formed is $Al_2Zn_3$, its calculating model of mass action concentrations is the same as that of Cu–Pb melts, and the mass action concentrations can be evaluated with equations (5) and (7). Substituting the measured activities from reference [4] into equation (7), after regression gives $K_{Al\,Zn_3}=0.113\,393$. Inserting $K_{AL,Zn_3}$ into equation (5) gives the calculated mass action concentrations compared with the measured activities as shown in **figure 3**(a). It can be seen from the figure that except those points at $\Sigma x_{Zn}\geqslant 0.662$, where the melts are saturated with Zn, the rest calculated values basically confirm with the measured ones, testifying that equations (5) and (7) can reflect the structural reality of the melts at 653 K. But at 1 000 K, as shown in **table 4**, the measured activities of these melts not only exhibit positive deviation relative to Raoult's law, but also at $\Sigma x_{Zn}=0.5$, their $\Delta G^m$ have minimum values, and $\Delta G^{xs}$ maximum values. Judging from the consistency of thermodynamic properties with the structure of the melts, It is conjectured that there should be a AB form compound formed in the melts. Hence the structural units of this two phase solution are Al, Zn atoms and AlZn compound (or short-range order chemical cluster), and form two solutions Al+AlZn and Zn+AlZn. From this, assuming the composition of the melts as $a=\Sigma x_{Zn}$, $b=\Sigma x_{AL}$, the equilibrium mole fraction of every structural unit expressed by the composition of the melts as $x=x_{Al}$, $y=x_{Zn}$, $z=x_{AlZn}$, and the mass action concentration of every structural unit as $N_1=N_{Al}$, $N_2=N_{Zn}$, $N_3=N_{AlZn}$ then it gives chemical equilibrium:

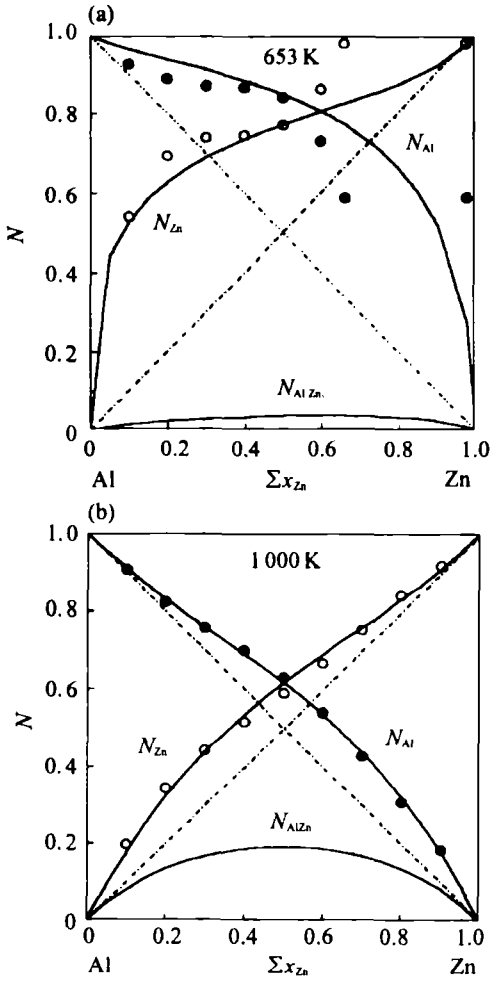


**Figure 3 Comparison of calculated mass action concentrations(—) with measured activities (• ○) for Al–Zn melts at different temperatures, (a) 653K; (b) 1 000 K.**

$$Al_{(l)}+Zn_{(l)}=AlZn_{(l)} \qquad K=\frac{N_3}{N_1N_2} \tag{15}$$

Mass balance:

$$b=x+z,\ a=y+z \tag{16}$$

$$N_1=1/b,\ N_2=1/a,\ N_3=KN_1N_2 \tag{17}$$

Substituting equation (17) into equation (16) gives:

$$N_1+KN_1N_2/b=1,\ N_2+KN_1N_2/a=1 \tag{18}$$

Summation of equation (18) gives:

$$K=ab(2-N_1-N_2)/[(a+b)N_1N_2] \tag{19}$$

The equations (18) and (19) are the calculating model of mass action concentrations for these melts at 1 000 K, in which equation (18) is used to calculate the mass action concentrations, while equation (19) for evaluation of equilibrium constant. Substituting the measured activities from reference [4] into equation (19) gives $K_{AlZn}=0.505\,069$, and inserting $K_{ALZN}$ into equation (18) gives the comparison of calculated mass action concentrations with measured activities as shown in figure 3 (b). It can be seen from the figure that the agreement of calculated and measured values is quite well, showing that the model deduced can exactly illustrate the structural reality of these melts at 1 000 K. From this paragraph, it is clear that at lower temperature there is a comparative complex compound ($Al_2Zn_3$) formed, while at higher temperature a relatively simple compound (AlZn) formed in these melts. This further confirms that these metastable compounds are not stable indeed.

**Table 4 Measured activities as well as $\Delta G^m$ and $\Delta G^{xs}$ of Al-Zn melts at 1 000 K**

| $\Sigma x_{Zn}$ | $a_{Al}$ | $a_{Zn}$ | $\Delta G^m$/(J·mol$^{-1}$) | $\Delta G^{xs}$/(J·mol$^{-1}$) |
|---|---|---|---|---|
| 0.1 | 0.905 | 0.197 | −2 101.77 | 602.90 |
| 0.2 | 0.824 | 0.344 | −3 064.74 | 1 101.13 |
| 0.3 | 0.759 | 0.443 | −3 638.33 | 1 440.26 |
| 0.4 | 0.699 | 0.516 | −3 985.83 | 1 611.92 |
| 0.5 | 0.629 | 0.588 | −4 149.93 | 1 624.48 |
| 0.6 | 0.539 | 0.666 | −4 086.32 | 1 511.43 |
| 0.7 | 0.428 | 0.753 | −3 772.31 | 1 310.47 |
| 0.8 | 0.308 | 0.840 | −3 123.35 | 1 038.33 |
| 0.9 | 0.184 | 0.916 | −2 059.91 | 644.77 |

## 5 Ga–Cd melts

The measured activities of Ga–Cd melts at 700 K as well as $\Delta G^m$ and $\Delta G^{xs}$ are given in **table 5** [4]. It can be seen from the table that activities of these melts exhibit positive deviation with respect to Raoultian behavior, testifying that they are two phase solution indeed, and at $\Sigma x_{Cd}=0.5$, $\Delta G^m$ has minimum value, $\Delta G^{xs}$ exhibits maximum value, but both activities and thermodynamic parameters haven't any obvious symmetrical character. Judging from the consistency of thermodynamic properties with the structure of the melts, except

**Table 5 $\Delta G^m$ and $\Delta G^{xs}$ of Ga–Cd melts at 700 K**

| $\Sigma x_{Cd}$ | $a_{Ga}$ | $a_{Cd}$ | $\Delta G^m$/(J·mol$^{-1}$) | $\Delta G^{XS}$/(J·mol$^{-1}$) |
|---|---|---|---|---|
| 0.1 | 0.445 | 0.940 | −858.29 | 1 034.14 |
| 0.2 | 0.606 | 0.890 | −1 126.25 | 1 787.76 |
| 0.3 | 0.714 | 0.844 | −1 260.23 | 2 298.55 |
| 0.4 | 0.770 | 0.811 | −1 323.03 | 2 595.82 |
| 0.5 | 0.803 | 0.784 | −1 348.15 | 2 687.93 |
| 0.6 | 0.820 | 0.764 | −1 339.78 | 2 579.07 |
| 0.7 | 0.843 | 0.724 | −1 276.97 | 2 277.62 |
| 0.8 | 0.876 | 0.645 | −1 126.25 | 1 787.76 |
| 0.9 | 0.924 | 0.468 | −799.68 | 1 096.94 |

AB form metastable compound, there should be other compound accompanying it. After repeated examination, it has been found that the calculating model considering both GaCd and $Ga_2Cd_3$ agrees best with practice. Hence, the structural units of the two phase solution Ga–Cd are Ga, Cd atoms as well as GaCd and $Ga_2Cd_3$ metastable compounds. Assuming the composition of the melts as $a=\Sigma x_{Cd}$, $b=\Sigma x_{Ga}$, the equilibrium mole fraction of every structural unit expressed by the composition of the melts as $x=x_{Ga}$, $y=x_{Cd}$, $z_1=x_{GaCd}$, $z_2=x_{Ga_2Cd_3}$, and the mass action concentration of every structural unit as $N_1=N_{Ga}$, $N_2=N_{Cd}$, $N_3=N_{GaCd}$, $N_4=N_{Ga_2Cd_3}$, then it gives chemical equilibrium:

$$Ga_{(l)}+Cd_{(l)}=GaCd_{(l)} \qquad K_1=\frac{N_3}{N_1N_2} \tag{20}$$

$$2Ga_{(l)}+3Cd_{(l)}=Ga_2Cd_{3(l)} \qquad K_2=\frac{N_4}{N_1^2N_2^3} \tag{21}$$

Mass balance:

$$b=x+z_1+2z_2 \tag{22}$$

$$a=y+z_1+3z_2 \tag{23}$$

$$N_1=x/b,\ N_2=y/a,\ N_3=K_1N_1N_2,\ N_4=K_2N_1^2N_2^3 \tag{24}$$

Substituting equation (24) into equations (22) and (23) gives:

$$\begin{cases} N_1+(K_1N_1N_2+2K_2N_1^2N_2^3)/b=1 \\ N_2+(K_1N_1N_2+3K_2N_1^2N_3^3)/a=1 \end{cases} \tag{25}$$

Summation of equation (25) gives:

$$ab(2-N_1-N_2)=K_1(a+b)N_1N_2+K_2(2a+3b)N_1^2N_2^3 \tag{26}$$

The equations (25) and (26) are the calculating model of mass action concentrations for Ga–Cd two phase melts, in which equation (25) is used for calculation of mass action concentrations, while equation (26) for evaluation of equilibrium constant.

Using measured activities of Ga–Cd melts at 700 K [4] and regressing by equation (26) gives $K_{GaCd}(=K_1)$= 0.138 786 5 and $K_{Ga_2Cd_3}(=K_2)$=0.002 761 9, then inserting both equilibrium constants into equation (25)gives the comparison of calculated mass action concentrations with measured activities as shown **figure 4**. It is clear from the figure that the agreement between calculated and measured values is satisfactory, testifying that the calculating model deduced can reflect the structural reality of these melts.

From the above discussions it is clear that binary metallic melts involving monotectic in the majority cases form between two elements have relatively great difference on their atomic mass. The difference between binary metallic melts involving monotectic and eutectic is that in the eutectic case, two solutions are mechanically mixed together, macroscopically one cannot distinguish both of the two solutions, while in the monotectic case, the melts are macroscopically separated into two layers. The metastable compounds formed in this type of binary metallic melts may be $A_2B_3$, $AB_2$, $A_2B_3$ or AB as well as $A_2B_3$+AB etc. There is a tendency that comparative complex compound formed, at lower temperature will change to relatively simple compound at higher temperature. As this type of melts is macroscopical two solutions, due to the resistance of diffusion across the boundary layer, the difficulty for

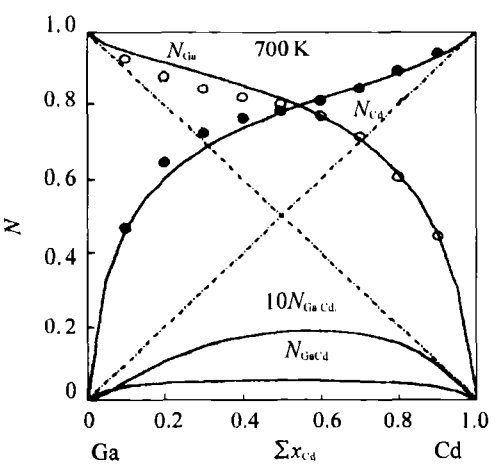


**Figure 4 Comparison of calculated mass action concentrations(—) with measured activities (•○) for Ga–Cd melts at 700 K.**

two elements to reach equilibrium is greater than that in case of binary metallic melts involving eutectic. Finally, the activities of two elements constituting this type of binary metallic melts generally exhibit positive deviations relative to Raoult's law.

## 6 Conclusions

(1) Based on the distinct immiscibility of two liquids in the binary phase diagrams of metallic alloy, measured activities as well as the behaviors of $\Delta G^m$ and $\Delta G^{ex}$, calculating models of mass action concentrations for binary metallic melts involving monotectic have been formulated. The calculated results agree with practice on the whole, showing that the models deduced basically can reflect the structural reality of this type of melts.

(2) Binary metallic melts involving monotectic in the majority cases form between two elements having relative great difference on their atomic mass. The difference between binary metallic melts involving monotectic and those involving eutectic is that in the latter case, two solutions are mechanically mixed together, macroscopically one cannot distinguish both of the two solutions, while in the former case, the melts are macroscopically separated into two layers.

(3) The metastable compounds formed in this type of binary metallic melts may be $A_2B_3$, $AB_2$, $A_2B_3$ or AB as well as $A_2B_3$+AB etc. There is a tendency that comparatively complex compound formed at lower temperature will change to relatively simple compound at higher temperature.

(4) As this type of melts is macroscopical two solutions, due to the resistance of diffusion across the boundary layer, the difficulty for two elements to reach equilibrium is greater than that in case of binary metallic melts involving eutectic.

(5) The activities of two elements constituting this type of binary metallic melts generally exhibit positive deviations relative to Raoult's law.

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