Study on the melting of interstitial alloy AB with FCC structure under pressure

Abstract

From the model of interstitial alloy AB with FCC structure and the condition of absolute stability for crystalline state we derive analytic expression for the temperature of absolute stability for crystalline state, the melting temperature and the equation of melting curve of this alloy by the way of applying the statistical moment method. The obtained results allow us to determine the melting temperature of alloy AB at zero pressure and under pressure. In limit cases, we obtain the melting theory of main metal A with FCC structure. The theoretical results are numerically applied for alloys AuSi and AgSi.

Keywords: interstitial alloy, absolute stability of crystalline state, statistical moment method

Introduction

Alloys in general and interstitial alloys in particular are typical materials in material technology and science. Study on interstitial alloys pays particular attention to many researchers. The melting temperature (MT) of materials under pressure is a very important problem of solid state physics and material science.1,3 Theoretically in order to determine the MT of crystal it is necessary to apply the equilibrium condition of solid phase and liquid phase. By this way, there are some methods such as the self-consistent phonon field method and the one-particle distribution function method. The obtained results from these methods are not in good agreement with experiments and are limited at low pressures.

In aid of the statistical moment method (SMM), Tang & Hung4,5 show that we absolutely only use the solid phase of crystal to determine the MT. The obtained results from the SMM are better than that from other methods in comparison with experiments.

Content of research

Analytic result

In the model of AB interstitial alloy with the face-centred cubic (FCC) structure, the A atoms with large size stay in the peaks and the face centers of cubic unit cell and the C interstitial atoms with smaller size stay in the body center. In1,2 we derived the analytic expressions of the nearest neighbor distance, the cohesive energy and the alloy parameters for atoms B, A, A1 (the atom A in the face centers) and A2 (the atom A in the peaks).

The equation of state of the AB interstitial alloy with FCC structure at temperature T and pressure P is described by

$$P_v = -\gamma_1 \left( \frac{1}{6} \frac{\partial u_b}{\partial r_b} + \theta x \cosh x + \frac{1}{2k} \frac{\partial k}{\partial r_b} \right)$$  \hspace{1cm} (1)

At 0K and pressure P, this equation has the form

$$P_v = -\gamma_1 \left( \frac{1}{6} \frac{\partial u_b}{\partial r_b} + \theta x \cosh x + \frac{1}{2k} \frac{\partial k}{\partial r_b} \right)$$  \hspace{1cm} (2)

Knowing the form of the interaction potential $\theta_{ij}$, equation (1) allows us to determine the nearest neighbor distance $r_{ij}(P,0)$ for each case of X. The displacement $\gamma_{0X}(P, T)$ of atoms from the equilibrium position at temperature T and pressure P is determined. From that, we can calculate the nearest neighbor distance $r_{ij}(P, T)$ at temperature T and pressure P as follows

$$\gamma_{ij}(P, T) = \gamma_{ij}(P,0) + \gamma_{ij}(P, T)$$

The mean nearest neighbor distance between two atoms in AB interstitial alloy with FCC structure is approximately determined by

$$\gamma_{ij}(P, T) = \gamma_{ij}(P,0) + \gamma_{ij}(P, T)$$

The free energy of AB interstitial alloy with FCC structure and concentration condition $c_b << c_a$ has the form

$$\psi_{AB} = (1 - 15c_a)\psi_a + c_b\psi_{ib} + 6c_b\psi_{ia} + 8c_b\psi_{ia} - T\gamma_c.$$

$$\psi_X \approx U_{0X} + \psi_{0X} + 3N \left\{ \frac{\beta^2}{k^2} \gamma_{2X}^2 \frac{X^2}{3} - \frac{2\gamma_{1X}}{3} \left( 1 + \frac{X^2}{2} \right) \right\} +$$

At 0K and pressure P, this equation has the form

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\[ P = \left( \frac{\partial \psi}{\partial V} \right)_T = -\frac{a}{3V} \left( \frac{\partial \psi}{\partial a} \right)_T. \]  

(6)

The pressure is calculated by

\[ \frac{a}{6} \left[ \frac{\partial \psi}{\partial a} \right] (1-15c_B) x_A \coth x_A + \frac{1}{k_B} \frac{\partial \psi}{\partial a} c_A x_A \coth x_A + \frac{1}{k_B} \frac{\partial \psi}{\partial a} x_A. \]  

(7)

where \( \psi \) is the free energy of \( A \) atom in pure metal, \( \psi_B \) is the free energy of \( B \) atom in interstitial alloy, \( \psi_A \) and \( \psi_B \) respectively are free energy of \( A \) and \( B \) atoms and \( S \) is the configuration entropy of \( AC \) interstitial alloy.

From the condition of absolute stability limit

\[ \frac{\partial P}{\partial V} = 0, \quad \frac{\partial P}{\partial a} = 0. \]  

(8)

Setting \( \gamma_G \) is the Grüneisen parameter of \( AB \) alloy. Then

\[ P = \frac{a}{6} \left[ \frac{\partial \psi}{\partial a} \right] (1-15c_B) x_A \coth x_A + \frac{1}{k_B} \frac{\partial \psi}{\partial a} c_A x_A \coth x_A + \frac{1}{k_B} \frac{\partial \psi}{\partial a} x_A. \]  

(9)

we can derive the absolute stability temperature for crystalline state in the form

\[ T_s = \frac{2 P S_B}{\gamma_G}, T_s = 2 P S_B + \frac{2}{6} \left[ (1-15c_B) \frac{\partial^2 U_{0,B}}{\partial a_B^2} + c_B \frac{\partial^2 U_{0,B}}{\partial a_B^2} + 6c_B \frac{\partial^2 U_{B,B}}{\partial a_B^2} + 8c_B \frac{\partial^2 U_{B,B}}{\partial a_B^2} \right] - \left[ (1-15c_B) \frac{\partial^2 U_{0,B}}{\partial a_B^2} + c_B \frac{\partial^2 U_{0,B}}{\partial a_B^2} + 6c_B \frac{\partial^2 U_{B,B}}{\partial a_B^2} + 8c_B \frac{\partial^2 U_{B,B}}{\partial a_B^2} \right] \]  

(10)

In the case of zero pressure,

\[ T_s = \frac{2 S_B}{6} \left[ (1-15c_B) \frac{\partial^2 U_{0,B}}{\partial a_B^2} + c_B \frac{\partial^2 U_{0,B}}{\partial a_B^2} + 6c_B \frac{\partial^2 U_{B,B}}{\partial a_B^2} + 8c_B \frac{\partial^2 U_{B,B}}{\partial a_B^2} \right] - \left[ (1-15c_B) \frac{\partial^2 U_{0,B}}{\partial a_B^2} + c_B \frac{\partial^2 U_{0,B}}{\partial a_B^2} + 6c_B \frac{\partial^2 U_{B,B}}{\partial a_B^2} + 8c_B \frac{\partial^2 U_{B,B}}{\partial a_B^2} \right] \]  

(11)

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\[ MS_2 = \left( 1 - 15c_B \right) \frac{a_{AB}^2 k_B}{4k_A^2} \left( \frac{\partial k_A}{\partial a_A} \right)^2 + c_B \frac{a_{AB}^2 k_B}{4k_B^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2 + 6c_B \frac{a_{AB}^2 k_B}{4k_B^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2 + \frac{a_{AB}^2 k_B}{4k_B^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2 + 8c_B \frac{a_{AB}^2 k_B}{4k_B^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2, \]

(11)

Because the curve of absolute stability limit for crystalline state is not far from the MS of crystal, the Ts temperature usually is large and \( x_s \) coth \( x_s \approx 1 \) at \( T_s \). Therefore,

\[ \frac{1}{1 - 15c_B} \frac{a_{AB}^2 k_B}{4k_A^2} \left( \frac{\partial k_A}{\partial a_A} \right)^2 + c_B \frac{a_{AB}^2 k_B}{4k_B^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2 + 6c_B \frac{a_{AB}^2 k_B}{4k_B^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2 + \frac{a_{AB}^2 k_B}{4k_B^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2 + 8c_B \frac{a_{AB}^2 k_B}{4k_B^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2 \times \]

\[ \times \left\{ 2PV_{AB} - \frac{a_{AB}^2 k_B}{4k_A^2} \left( 1 - 15c_B \right) \frac{\partial^2 U_{0,AB}}{\partial a_A^2} + c_B \frac{\partial^2 U_{0,AB}}{\partial a_B^2} + 6c_B \frac{\partial^2 U_{0,AB}}{\partial a_B^2} + 8c_B \frac{\partial^2 U_{0,AB}}{\partial a_B^2} - \left( 1 - 15c_B \right) \frac{a_{AB}^2 k_B}{2k_A c_B} \left( \frac{\partial k_A}{\partial a_A} \right)^2 + a_{AB} \left( \frac{\partial k_B}{\partial a_B} \right)^2 + c_B \left( \frac{\partial k_B}{\partial a_B} \right)^2 \right\} \]

\[ = -6c_B \left[ \frac{a_{AB}^2 k_B}{2k_A c_B} \left( \frac{\partial k_A}{\partial a_A} \right)^2 - a_{AB} \left( \frac{\partial k_B}{\partial a_B} \right)^2 - \frac{a_{AB}^2 k_B}{4k_A^2} \left( \frac{\partial k_B}{\partial a_B} \right)^2 \right] + \]

\[ + \left[ \frac{a_{AB}^2 k_B}{2k_A c_B} \left( \frac{\partial k_B}{\partial a_B} \right)^2 + c_B \frac{\partial^2 U_{0,AB}}{\partial a_B^2} + 6c_B \frac{\partial^2 U_{0,AB}}{\partial a_B^2} + 8c_B \frac{\partial^2 U_{0,AB}}{\partial a_B^2} \right] \times \]

\[ \left\{ \frac{1}{3} \frac{a_{AB}^2 k_B}{4k_A^2} \left( 1 - 15c_B \right) + \frac{1}{2k_A} \frac{\partial k_A}{\partial a_A} + \frac{1}{k_A} \frac{\partial k_A}{\partial a_A} - 6c_B + \frac{1}{k_A} \frac{\partial k_B}{\partial a_B} - 8c_B \right\} \]

\[ \times \left\{ P + \frac{a_{AB}^2 k_B}{6V_{AB}} \left( 1 - 15c_B \right) \frac{\partial U_{0,AB}}{\partial a_A} + c_B \frac{\partial U_{0,AB}}{\partial a_B} + 6c_B \frac{\partial U_{0,AB}}{\partial a_B} + 8c_B \frac{\partial U_{0,AB}}{\partial a_B} \right\} = 0. \]

(12)

That is the equation for the curve of absolute stability limit for crystalline state. Therefore, the pressure is a function of the mean nearest neighbor distance

\[ P = P(a_{AB}). \]

(13)

Temperature \( T_s \) at zero pressure has the form

\[ T_s = a_{AB} \frac{\partial U_{0,AB}}{\partial T} \left( \frac{\partial U_{0,AB}}{\partial T} \right)^T \gamma_G, \]

(14)

where the parameters \( a_{AB}, \frac{\partial U_{0,AB}}{\partial T}, \gamma_G \) are determined at \( T_s(0) \).

Temperature \( T_s \) at pressure \( P \) has the form

\[ T_s \approx T_s(0) + \frac{V_{AB}^2}{3k_B^2} \left( \frac{\partial U_{G}}{\partial T} \right)^T T_s. \]

(15)

Here, \( k_B \) is the Boltzmann constant, \( V_{ABC}, \gamma_G, \frac{\partial U_{G}}{\partial T} \) are determined at \( T_s \). Approximatel, \( T_s(0) \approx T_s \). In order to solve equation (15), we can use the approximate iteration method. In the first approximate iteration,

\[ T_{s1} = T_s(0) + \frac{V_{AB}}{3k_B^2} \frac{\partial U_{G}}{\partial T} T_s(0). \]

(16)

Here \( T_s(0) \) is the temperature of absolute stability limit for crystalline state at pressure \( P \) in the first approximate iteration of equation (15). Substituting \( T_{s1} \) into equation (15), we obtain the better approximate value \( T_{s2} \) of \( T_s \) at pressure \( P \) in the second approximate iteration

\[ T_{s2} \approx T_s(0) + \frac{V_{AB}}{3k_B^2} \frac{\partial U_{G}}{\partial T} T_s(0). \]

(17)

Analogously, we can obtain the better approximate values \( T_{s3}, T_{s4}, \ldots \) of \( T_s \) at pressure \( P \) in the third, fourth, etc. approximate iterations. These approximations are applied at low pressures.

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In the case of high pressure, the MT of alloy at pressure $P$ is calculated by

$$T_m(P) = \frac{T_m(0)B_0}{G(0)} \left( \frac{G(P)}{G(0)} \right)^{1/(B_0 + B'_0P)}$$

(18)

where $T_m(P)$ and $T_m(0)$ respectively are the MT at pressure $P$ and zero pressure, $G(P)$ and $G(0)$ respectively are the rigidity bulk modulus at pressure $P$ and zero pressure, $B_0$ is the isothermal elastic modulus at zero pressure, $B'_0 = \left( \frac{dB_0}{dP} \right)_{P=0}$, $B'_0(P)$ is the isothermal elastic modulus at pressure.

**Numerical results for alloys AuSi and AgSi**

For alloys AuSi and AgSi, we use the $n–m$ pair potential

$$\varphi(r) = \frac{D}{n - m} \left[ m \left( \frac{r_0}{r} \right)^n - n \left( \frac{r_0}{r} \right)^m \right]$$

(19)

where potential parameters are given in Table 1.

**Table 1 Potential parameters $m$, $n$, $D$, $r_0$ of materials**

| Material | $m$  | $n$  | $D \left[ 10^{-16} \text{ erg} \right]$ | $r_0 \left[ 10^{-10} \text{ m} \right]$ |
|----------|------|------|----------------------------------------|----------------------------------------|
| Au       | 5.5  | 10.5 | 6462.540                               | 2.8751                                 |
| Ag       | 5.5  | 11.5 | 4589.328                               | 2.8760                                 |
| Si       | 6.0  | 12.0 | 45128.340                              | 2.2950                                 |

Considering the interaction between atoms Au(Ag) and Si in the above mentioned alloys, we use the potential (19) but calculating approximately $\tilde{D} = \sqrt{D_{\text{AuAg}}D_{\text{Si}}}$, $\tilde{r}_0 = \sqrt{\tilde{r}_{\text{AuAg}}r_{\text{Si}}}$. Parameters $\tilde{m}$ and $\tilde{n}$ are taken empirically.

At 0.1 MPa, Au has a FCC structure with $a = 4.0785 \times 10^{-10} \text{ m}$ at 300K and the melting point at 1337 K. The melting curve of Au is determined up to 1673 K and 6.5 GPa with the slope $dT/dP = 60 \text{ K/ GPa}$, and up to 1923 K and 12 GPa.

At 0.1 MPa, Ag has a FCC structure with $a = 4.0862 \times 10^{-10} \text{ m}$ at 300K and the melting point at 1235 K. The melting curve of Ag is determined up to 1563 K and 6.5 GPa with the slope $dT/dP = 60 \text{ K/ GPa}$.

Our numerical results are summarized in tables from Table 2 & Table 3 and illustrated in figures from Figure 1 & Figure 2. The concentration of interstitial atoms changes from 0 to 5% and the pressure changes from 0 to 10 GPa for Au and from 0 to 6 GPa for Ag.

**Table 2 Dependence of melting temperature on pressure and concentration of interstitial atoms for alloy Au–xSi**

| $x$(%) | P(GPa) | 0      | 2      | 4      | 6      | 8      | 10     |
|--------|--------|--------|--------|--------|--------|--------|--------|
| 0      | SMM    | 1337.33| 1397.27| 1454.33| 1514.75| 1568.48| 1629.22|
|        | EXPT12 | 1337.33| 1432.90| 1530.15| 1628.63| 1727.10| 1824.36|
|        | EXPT14 | 1340.30| 1426.80| 1515.77| 1599.80| 1683.84| 1770.34|
| 1      | SMM    | 1382.99| 1443.15| 1500.10| 1560.32| 1613.50| 1673.78|
| 3      | SMM    | 1505.78| 1567.89| 1625.75| 1686.56| 1739.30| 1799.29|
| 5      | SMM    | 1701.80| 1771.20| 1834.58| 1900.23| 1955.89| 2018.94|

**Figure 1** Dependence of melting temperature on pressure and concentration of interstitial atoms for alloy AuSi.

**Figure 2** Dependence of melting temperature on pressure and concentration of interstitial atoms for alloy AgSi.

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According to our numerical results for alloy AuSi at the same concentration of interstitial atoms Si when pressure increases, the melting temperature increases. For example at $c_{Si} = 5\%$ when $P$ increases from 0 to 10 GPa, $T_{melting}$ of alloy AuSi increases from 1701.8 K to 2018.94 K. At the same pressure when the concentration of interstitial atoms Si increases, the melting temperature increases. For example at $P=10$ GPa, when $c_{Si}$ increases from 0 to $5\%$, $T_{melting}$ of alloy AuSi increases from 11629.22 K to 2018.94K.

According to our numerical results for alloy AgSi at the same concentration of interstitial atoms Si when pressure increases, the melting temperature increases. For example at $c_{Si} = 5\%$ when $P$ increases from 0 to 6 GPa, $T_{melting}$ of alloy AgSi increases from 1418.12 K to 1764.81 K. At the same pressure when the concentration of interstitial atoms Si increases, the melting temperature increases. For example at $P=6$ GPa, when $c_{Si}$ increases from 0 to $5\%$, $T_{melting}$ of alloy AuSi increases from 1492.95 K to 1764.81 K (see Table 3).

### Table 3 Dependence of melting temperature on pressure and concentration of interstitial atoms for alloy Ag–xSi

| $x(\%)$ | $P$(GPa) | 0     | 2     | 4     | 6     |
|--------|----------|-------|-------|-------|-------|
|        |         | 1234.93 | 1322.75 | 1408.68 | 1492.95 |
|        | SMM      | 1234.93 | 1334.42 | 1438.19 | 1538.81 |
|        | EXPT     | 1257.82 | 1349.70 | 1438.34 | 1524.38 |
|        | $c_{Si}=1\%$ | 1319.47 | 1423.32 | 1520.45 | 1612.43 |
|        | $c_{Si}=3\%$ | 1418.12 | 1544.47 | 1659.14 | 1764.81 |

At zero concentration of interstitial atoms Si, the melting temperature of alloys AuSi and AgSi respectively becomes the melting temperature of metals Au and Ag. The melting temperature of substitution alloys AuCu and AgCu is smaller than the melting temperature of metals Au and Ag. The melting temperature of alloys AuSi and AgSi respectively becomes the melting temperature of metals Au and Ag at zero pressure. According to our numerical results for alloy AgSi at the same concentration of interstitial atoms Si when pressure increases, the melting temperature increases. For example at $P=10$ GPa, when $c_{Si}$ increases from 0 to $5\%$, $T_{melting}$ of alloy AuSi increases from 11629.22 K to 2018.94 K. At the same pressure when the concentration of interstitial atoms Si increases, the melting temperature increases. For example at $P=10$ GPa, when $c_{Si}$ increases from 0 to $5\%$, $T_{melting}$ of alloy AuSi increases from 11629.22 K to 2018.94 K (see Table 3).

### Table 4 The melting temperatures of metals Au and Ag at zero pressure

| Metal          | Au       | Ag       |
|----------------|----------|----------|
| $T_m$ (K)      | 1400     | 1190     |
| SMM            | 1336     | 1234     |
| EXPT           |          |          |

### Table 5 The melting temperatures of metals Au and Ag under pressure

| P(GPa) | Method | 1     | 2     | 3     | 4     | 5     | 6     |
|--------|--------|-------|-------|-------|-------|-------|-------|
|        | SMM    | 1476  | 1543  | 1602  | 1651  | 1693  | 1728  |
|        | EXPT   | 1383  | 1455  | 1513  | 1575  | 1625  | 1667  |
| Au     |        | 1282  | 1367  | 1443  | 1513  | 1575  | 1631  |
|        | SMM    | 1300  | 1353  | 1403  | 1462  | 1509  | 1549  |
| Ag     |        | 1234  | 1319  | 1394  | 1463  | 1528  | 1583  |

### Conclusion

In aid of the SMM, we derive the analytic expressions of the temperature of absolute stability limit for crystalline state and the melting temperature together with the melting curve of the binary interstitial alloy depending on pressure and concentration of interstitial atoms. In limit case, we obtain the melting theory of main metal with FCC structure. The theoretical results are numerically applied for alloys AuSi and AgSi.

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### Conflict of interest

Author declares there is no conflict of interest.

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