Investigation of ordering phenomenon in Me–Pt (Me = Fe,Ni) liquid alloys

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Abstract
The phase diagrams of Fe–Pt and Ni–Pt liquid alloy systems show the existence of FePt and NiPt intermetallic compounds, respectively, in their solid intermediate states, and the associative tendency between unlike atoms in these liquid alloys has been analysed using the self-association model. The concentration dependences of mixing properties such as the free energy of mixing, \(G_M\); the concentration fluctuations, \(S_{cc}(0)\), in the long-wavelength limits; the chemical short-range order (CSRO) parameter, \(\alpha_1\); as well as the chemical diffusion, enthalpy and entropy of the mixing of Fe–Pt and Ni–Pt liquid alloys have been investigated to determine the nature of ordering in the liquid alloys. The results show that heterocoordination occurs in the alloys at all concentrations. The effect of CSRO on \(S_{cc}(0)\), chemical diffusion, \(D\), and the order parameter, \(\alpha_1\), has been considered. The ordering phenomenon in the liquid alloys is also related to the effect of the atomic size mismatch volume on \(S_{cc}(0)\).

Keywords: intermetallic, raoultian behaviour, size mismatch, concentration fluctuations

1. Introduction
A number of theoretical models have been proposed by theoreticians, physicists, chemists and metallurgists to explain the concentration dependence of the thermodynamic functions of binary liquid alloys, and thus, extract useful microscopic information on them [1–5]. Many investigations have been reported in the literature on liquid binary alloys, which are of importance from both scientific and technological points of view. Accurate knowledge of the mixing properties and phase diagrams of the alloy systems is essential for establishing a good understanding of the relationship between the experimental results, theoretical approaches and empirical models for liquid alloys.

The mixing behaviour of two metals forming binary alloys is the result of the interplay between the energetic and structural readjustment of the constituent elemental atoms. Upon alloying, A and B atoms can remain self-coordinated, forming A–A or B–B pairs, or can exhibit a strong interactive tendency between unlike atoms, forming heterocoordinated A–B pairs. Alloys of the type \(M_\alpha A_m\), where \(M\) and \(A\) are metals or semimetals from different parts of the periodic table with chemical valences \(m\) and \(\alpha\), respectively, are often referred to as liquid alloys. Systems such as Au–Bi, Bi–Cd, Al–Bi, Ni–Pt and Fe–Pt are examples of liquid alloys. All liquid binary alloys can be classified according to the deviations of their thermodynamic and thermophysical functions from Raoultian behaviour into two main groups: segregating (positive deviation) or short-range ordered (negative deviation) alloys [6–12].

A number of intermetallics have attracted particular interest in recent years. Intermetallics, i.e. intermetallic compounds and ordered intermetallic phases, are a fascinating class of materials, from the points of view of both their fundamental properties and their practical applications [13]. For example, the bimetallic catalysts of Pt–Zn show anomalous catalytic functions for the hydrogenolysis of esters [14, 15], the hydrogenation of dienes, the conversion of unsaturated aldehydes to unsaturated alcohols and the partial oxidation of methanol [16]. Although little is known about the uses of platinum-based alloys, since iron, nickel and platinum are transition metals with nickel and platinum belonging to the same group of the periodic table (which implies that they possess similar chemical properties), it is likely that Ni–Pt and Fe–Pt will have similar catalytic properties to various applications of Pt–Zn.
The present interest in liquid Fe–Pt and Ni–Pt is twofold. Firstly, apart from the high academic interest, the wide diversity of their physical properties is of great commercial significance. Fe–Pt alloys can be used to monitor redox conditions in experiments including, those on Fe-bearing phases [17, 18]. Iron is a ubiquitous element within the Earth and other terrestrial planets and the search continues for suitable container materials for experimentation involving Fe-bearing samples [18]. Some prospective materials are limited by their low melting points (e.g. Ag, Au) and/or their tendency to react with the samples (e.g. [SiO₂, Al₂O₃]), whereas others can only be used under relatively reducing conditions (e.g. Mo, graphite). In many respects, Pt is an ideal container material; it can be used at high temperatures (e.g. 1600°C) and it is essentially inert. Secondly, the thermodynamic data obtained from experiments coupled with the constituent elements serve as guiding principles for the selection of Pt alloys studied in this work. In addition, the thermodynamic assessment of Fe–Pt systems has been performed using the CALPHAD approach (CALculation of PhAsse Diagram) by Fredriksson and Sundman [19], while the interaction parameter of the liquid phase in Ni–Pt has been critically evaluated in [20].

The knowledge of the structural, thermodynamic and thermophysical properties is essential for understanding the nature of ordering in liquid alloys. The associative tendency between unlike atoms in Fe–Pt and Ni–Pt alloys is strong, and therefore we have used the self-association model (SAM) [7, 10, 21] here. Various mixing properties such as the concentration dependence of the free energy of mixing, \( G_M/RT \); the thermodynamic activity, \( a_i \); the concentration fluctuations, \( S_i(0) \), in the long-wavelength limit; the Warren–Cowley chemical short-range order (CSRO) parameter, \( a_i \); and the chemical diffusion, \( D \), have been observed to deviate negatively from the ideal results, and thus, both alloys belong to the class of liquid alloys that exhibits a preference towards heterocoordination (short-range ordering). In addition, the enthalpy of mixing, \( H_M \), and the entropy of mixing, \( S_M \), of Fe–Pt and Ni–Pt molten alloys have also been evaluated. The results indicate that the enthalpies of mixing for the alloys are highly symmetric and negative (typical of ordered systems), while the entropies of mixing for the two alloys are symmetrical and positive at all concentrations, indicating atomic order.

In section 2, an overview of the SAM is given. This is followed in section 3 by results and discussion for various thermodynamic functions investigated using the framework of the SAM. Section 4 deals with the results for chemical diffusion, while the results for the enthalpy and entropy of mixing are discussed in section 5. A summary is given in section 6.

### 2. Model description

A simple scheme proposed by Singh and Sommer [11] for demixing liquid alloys, which has been successfully used to study binary liquid alloys, is applied to Fe–Pt and Ni–Pt alloys. Relevant applications of the model can be found in [7, 10, 21]. In the model, it is first assumed that the binary alloy consists of \( N_A = x_A N \) atoms of A and \( N_B = x_B N \) atoms of element B, so that the total number of atoms, \( N \), is \( N = N_A + N_B \). Here \( x_A \) is the mole fraction of A in the alloy. In addition, the SAM is based on the assumption that the atoms of the constituent elements A and B exist in the form of a polyatomic matrix, leading to the formation of like-atom clusters or self-associates of the type \( A_μ \) and \( B_ν \), i.e.

\[
\mu A = A_μ, \quad ν B = B_ν, \quad (1)
\]

where \( μ \) and \( ν \) are the numbers of atoms in the clusters of type A and type B matrices, respectively. The thermodynamic properties of the demixing liquid alloys are dependent on the number of self-associates; \( n = \frac{N}{μ} \). Thus, we make the following two assumptions: (i) all the atoms are located on a set of equivalent lattice sites with each having \( Z \) nearest neighbours; (ii) the interaction is short range and effective only between nearest neighbours. Using the Flory approximation [2, 11] (i.e. \( Z \to \infty \)), a simple relation for the Gibbs free energy of mixing is expressed as

\[
G_M = \frac{1}{RT} \left\{ x_A \ln x_A + (1 - x_A) \ln (1 - x_A) + x_A (1 - β) + \ln γ \right\} + x_A (1 - x_A) γ W \quad (2)
\]

with

\[
W = µ w, \quad (3)
\]

\[
n = \frac{µ}{ν}, \quad (4)
\]

\[
β = 1 - \frac{1}{n}, \quad (5)
\]

\[
γ = \frac{1}{1 - x_A β}, \quad (6)
\]

where \( w \) is the ordered energy or the interchanged energy, which by definition have the same physical meaning in this study:

\[
w = Z \left( ε_{AB} - (ε_{AA} + ε_{BB})/2 \right), \quad (7)
\]

where \( ε_{ij}, (i, j = A, B) \) denotes the energy of the nearest-neighbour \( i-j \) bond and \( Z \) is the coordination number of the liquid alloy. The model parameters \( n \) and \( W \) are independent of concentration but may depend on pressure \( P \) and temperature \( T \).

Equation (2) can be used to obtain an expression for activity \( a_i \) (\( i = A, B \)), i.e.

\[
RT \ln a_i = \left( \frac{\partial G_M}{\partial N_i} \right)_{T,P,N}. \quad (8)
\]

Recalling that \( N = N_A + N_B \) with \( x_A = N_A/N \) and substituting equation (2) in equation (8), we obtain the activities as

\[
\ln a_A = \ln \left[ x_A γ (1 - β) \right] + (1 - x_A) γ β + (1 - x_A)^2 γ^2 \frac{W}{RT} \quad (9)
\]
\[ \ln a_B = \ln(x_A \gamma) + x_A(1 - \beta)\gamma(1 - n) + n x_A^2(1 - \beta)\gamma^2 \frac{W}{RT}. \] 

Once the expressions for \( G_M \) and activity \( a_i (i = A, B) \) are known, other thermodynamic functions simply follow.

The entropy of mixing \( S_M \) is obtained from the thermodynamic relationship

\[ S_M = R \left( x_A \ln x_A - (1 - x_A) \ln(1 - x_A) \right) \]

and is given as

\[ S_M = x_A(1 - x_A) y' W - x_A(1 - x_A) y' T \frac{\partial W}{\partial T} + RT^2 x_A(1 - x_A) y' \left[ \frac{\beta}{1 - \beta} - x_A y' \frac{W}{RT} \right] \frac{\partial \beta}{\partial T}. \]

The concentration fluctuations in the long-wavelength limit, \( S_{cc}(0) \), can be easily calculated from the standard relationship in terms of the free energy of mixing,

\[ S_{cc}(0) = RT \left( \frac{\partial^2 G_M}{\partial x_A^2} \right)_{T,P,N}^{-1}. \]

Using equations (2) and (9) or (10), we determined \( S_{cc}(0) \) as

\[ S_{cc}(0) = \frac{x_A(1 - x_A)}{1 - x_A(1 - x_A) g(n, W)}. \]

where

\[ g(n, W) = \frac{2n^2(W/RT) - (n - 1)^2[(x_A + n(1 - x_A)]}{[(x_A + n(1 - x_A)]^3}. \]

Using equations (16), then all other parameters can be calculated. Thus, equations (2), (12), (13) and (16) are the essential equations for this investigation.

Table 1. Fitted interaction parameters for Fe–Pt and Ni–Pt liquid alloys

| Alloy   | \( T \) (K) | \( Z \) | \( W/RT \) | \( \partial W/\partial T \) | \( \partial \beta/\partial T \) | \( \mu \) | \( \nu \) |
|---------|-------------|--------|------------|-----------------------------|-------------------------------|-------|-------|
| Fe–Pt   | 1123        | 10     | -7.21      | 0.0001                      | 0                             | 1     | 1     |
| Ni–Pt   | 1625        | 10     | -5.80      | 0.00000                     | 1                             | 1     | 1     |

Figure 1. Concentration dependence of \( G_M^\text{Fe-Pt} \) for Fe–Pt liquid alloys at 1123 K. The solid line represents theoretical values and the crosses represent experimental values. \( c_{Pt} \) is the Pt concentration in the alloy. The experimental data are from [22].

3. Results and discussion

3.1. Free energy of mixing and activity

The most important task in performing the numerical calculation is to fix the number of atoms \( \mu \) and \( \nu \) of the complexes (\( \mu \text{Fe} + \nu \text{Pt} = \text{Fe}_\mu\text{Pt}_\nu \)) and (\( \mu \text{Ni} + \nu \text{Pt} = \text{Ni}_\mu\text{Pt}_\nu \)). From the phase diagram [22], it is clear that Fe–Pt and Ni–Pt can exist as intermetallic compounds in the solid state, FePt (\( \mu = 1, \nu = 1 \)) and NiPt (\( \mu = 1, \nu = 1 \)) at 1123 and 1625 K, respectively, suggesting short-range ordering in the liquid phase [23]. Of course, it is not unlikely for such associates to exist in some form in the liquid phase close to the melting temperature. Once \( \mu \) and \( \nu \) are obtained, then equation (2) can be utilized to determine the optimal values of the interaction parameters that give the best representation of the experimental Gibbs free energy of mixing, \( G_M \). Table 1 shows the values of the fitted interaction parameters for the liquid Fe–Pt and Ni–Pt alloys. While keeping these fitted parameters invariant, one can use them to compute \( S_{cc}(0), a_i \) and \( a_1 \), and thus, form a basis for explaining the energetics of the liquid alloys. We have used equation (2) to calculate \( G_M \) for both systems, while the experimental data were taken from [22]. The computed and experimental values of \( G_M^\text{Fe-Pt} \) as a function of composition are given for the two alloys in figures 1 and 2, respectively. The results suggest that both systems, Fe–Pt and Ni–Pt, with \( G_M^\text{min} \) values of \(-2.496\) \( RT \) and \(-1.388\) \( RT \), respectively, are less strongly interacting than Mg–Bi (\( G_M^\text{min} = [-3.38\) \( RT \)) and liquid amalgams such as Hg–K (\( G_M^\text{min} = [-3.35\) \( RT \)) and Hg–Na (\( G_M^\text{min} = [-3.23\) \( RT \)) [24]. From a close examination of the figures, it is observed that our energy parameters give a good representation of the experimental free energy of mixing for the alloys studied. The calculated activities obtained using
equation (9) or (10) are compared with the experimental activities in table 2. The results presented in table 2 show that there is good agreement between the experimental and calculated activities of Ni–Pt, while the experimental activity for Fe–Pt is not available at the temperature of interest. The excellent fits obtained for $G_M$ and the results for the thermodynamic activity allow the confident use of the fitted interaction parameters to study the nature of ordering in the liquid alloys.

### 3.2. Concentration fluctuations. Warren–Cowley CSRO parameter and ordered energy

$S_{cc}(0)$ is a significant microscopic parameter in understanding the nature of atomic order in binary liquid alloys [11, 25]. It is also directly related to thermophysical properties such as chemical diffusion [26–28]. Once the Gibbs free energy of mixing, $G_M$, of a liquid phase is known, $S_{cc}(0)$ can be expressed in terms of $G_M$, or in terms of activity, $a_i$ as in equations (14) and (15). In principle, $S_{cc}(0)$ can be determined directly from small-angle diffraction experiments, but because of the difficulty of the experimental procedure it has never been accomplished successfully. In contrast, the theoretical calculation of $S_{cc}(0)$ is of considerable importance when the nature of atomic interactions in the melt has to be analysed. The experimental values of $S_{cc}(0)$ for the two systems in this study were obtained by numerical differentiation of the experimental free energy of the mixing equation (14)
with that of FePt (~1310 °C = 1583 K). This is in contrast to the Ni–Pt system, which has almost the same order–disorder phase transition temperatures for Ni3Pt (~580 °C = 853 K) and NiPt (~645 °C = 918 K).

The effect of atomic size mismatch volume on \( S_{cc}(0) \) in the long-wavelength limit is also considered. The size ratio is \( \Omega_A, \Omega_B > \Omega, \) where \( \Omega \) is the atomic volume. It is clear from equation (16) that the values of \( S_{cc}(0) \) due to the size mismatch volumes (\( \Omega_A = 7.94, \Omega_B = 7.43, 10.31, \) where \( j = Fe, Ni, Pt, \) respectively) are always less than the ideal values, which suggests chemical order in the alloys. It must, however, be noted that for \( \Omega_A \neq \Omega_B, \) \( S_{cc}(0) \) deviates from the ideal value, whereas if \( \Omega_A = \Omega_B, \) then \( S_{cc}(0) = S_{cc}^{id}(0) = x_A x_B. \) Thus, any positive deviation of \( S_{cc}(0) \) from the ideal value, i.e., segregation, should not be attributed to the atomic size mismatch volume alone.

To obtain a clear picture of the nature of segregation and ordering in the binary liquid alloys, another quantity known as the Warren–Cowley CSRO parameter, \( \alpha_1, [6, 24, 29] \) is computed. This is related to the \( S_{cc}(0) \) by

\[
\alpha_1 = \frac{(S - 1)}{S(Z - 1) + 1},
\]

where

\[
S = \frac{S_{cc}(0)}{x_A (1 - x_A)}.
\]

For uniformity, we have chosen \( Z \) as 10 in the liquid phase [8, 12, 30]. Note that the choice of \( Z \) does not significantly affect our results. For equiatomic composition, the CSRO parameter, \( \alpha_1, \) is found to be \(-1 \leq \alpha_1 \leq 1\). Negative values of this parameter indicate ordering in the melt, and complete ordering is indicated by \( \alpha_1^{\text{min}} = -1. \) In contrast, positive values of \( \alpha_1 \) indicate segregation, whereas phase separation takes place if \( \alpha_1^{\text{max}} = 1. \) The variation of \( \alpha_1 \) as a function of concentration is shown in figure 5 for the two liquid alloys. It is seen that \( \alpha_1 \) is negative across the whole concentration range, as expected for ordered systems. This signifies that the two alloys studied are heterocoordinated systems, as evident from the calculations of \( \frac{\partial S}{\partial T} \) and \( S_{cc}(0). \) However, the degree of order is greater in Fe–Pt (\( \alpha_1^{\text{min}} = -0.265 \)) at \( c_{Pt} = 0.5 \) than in Ni–Pt (\( \alpha_1^{\text{min}} = -0.114 \)) at \( c_{Pt} = 0.5. \) Also, \( \alpha_1 \) is symmetrical about the equiatomic composition.

Table 1 shows the values of the parameters used for the calculations. Generally, if the ordered energy, \( \omega, \) is less than zero, there is a tendency to form unlike-atom pairs, and if \( \omega > 0, \) then like atoms tend to pair together. However, \( \omega = 0, \) indicating that atoms in the mixtures are perfectly disordered [1]. Hence, the fact that \( \omega < 0 \) for Fe–Pt and Ni–Pt (table 1) shows that there is a greater tendency for unlike atoms to pair in the two alloys, which implies heterocoordination. In addition, \( \omega = -7.21 \) and \( \omega = -2.58 \) for Fe–Pt and Ni–Pt, respectively, which further confirms that Fe–Pt is more highly interacting than Ni–Pt.

4. Chemical diffusion

The knowledge of \( S_{cc}(0) \) has been further used to investigate the chemical diffusion in the two liquid alloys, which is likely to play an important role in many technological and corrosion phenomena. Using the Darken thermodynamic equation for diffusion, an expression that relates diffusion and \( S_{cc}(0) \) can be established [6, 9, 27]; thus, one can write

\[
\frac{D_M}{D_{id}} = \frac{S_{cc}^{id}(0)}{S_{cc}(0)},
\]

where \( D_M \) is the mutual diffusion coefficient and \( D_{id} \) is the intrinsic diffusion coefficient for an ideal mixture, given as

\[
D_{id} = x_A D_A + x_B D_B.
\]

with \( D_A \) and \( D_B \) being the self-diffusion coefficients of pure components A and B, respectively. For ideal mixing, \( S_{cc}(0) \rightarrow S_{cc}^{id}(0), \) i.e., \( D_M \rightarrow D_{id}; \) for ordered alloys, \( S_{cc}(0) < S_{cc}^{id}(0), \) i.e., \( D_M > D_{id}; \) and similarly for segregation, \( D_M < D_{id}. \) The maximum peak of \( \frac{D_M}{D_{id}} \) as a function of composition indicates the maximum chemical order in a molten alloy system as well as the composition of the most probable associates to be formed in the liquid phase [23, 27]. The computed values of \( S_{cc}(0) \) have been used in equation (21) to evaluate \( \frac{D_M}{D_{id}} \) as a function of composition, as shown in figure 6. It is observed that \( \frac{D_M}{D_{id}} \) is greater than 1 in the entire concentration range for these alloys. This is indicative of the presence of chemical order in the alloys, and further corroborates our earlier hypothesis that the degree of order in Fe–Pt is higher than that in Ni–Pt.
5. Enthalpy and entropy of mixing

It can be seen from equation (11) for $H_M$ that to obtain a good fit to this parameter, we need to incorporate the temperature dependence of the interaction parameters; any other approximation would be invalid. Using equations (12) and (13), we have ascertained the variation in temperature parameters from the measured values of $H_M$ and $S_M$; the results are shown in table 1. From the fitted enthalpy of mixing and entropy of mixing, shown in figures 7 and 8, respectively, as functions of concentration, one observes that the fit obtained for the Ni–Pt alloy compared well with the experimental data, while the fit could not be compared for the Fe–Pt alloy due to the lack of experimental data at the temperature of interest for both the enthalpy and the entropy of mixing. The values obtained (table 1) show that the temperature dependence of the energy parameters is small and that the enthalpy of mixing for Fe–Pt and Ni–Pt alloys is highly symmetric and negative (typical of ordered systems), in agreement with the free energy of mixing, $G_M$, and the enthalpy of mixing, $H_M$, which exhibit negative deviations from Raoultian behaviour for the investigated alloys.

The entropy of mixing (figure 8) for the alloys is symmetrical and positive at all concentrations, which indicates atomic order. Generally, one observes that both the data reported in the literature and the theoretical values obtained using the interaction parameters and their temperature dependence are in good agreement.

6. Summary

In the present study, it has been shown that the two liquid alloys investigated are characterized by negative interaction energies, which implies that the alloys are short-range ordered alloys. The energetics of Fe–Pt and Ni–Pt molten alloys have been evaluated, focusing on their thermodynamic functions. The study of the concentration dependence of thermodynamic functions such as the free energy of mixing, concentration fluctuations, the Warren–Cowley CSRO parameter, the chemical diffusion and the enthalpy and entropy of the mixing of the liquid alloys have been used to deduce the nature of ordering in the melts. The associative tendency between the constituent elemental atoms of the liquid alloys has been analysed by the self-association model. Results on concentration fluctuations, $S_c(0)$, which are very useful for inferring the nature of atomic order in liquid alloys, have been described, and indicate heterocoordination in the liquid alloys across the entire range concentrations.

The degree of order in the investigated liquid alloys has been quantified using the CSRO parameter ($\alpha_1$), which is plotted as a function of concentration. Similar to $S_c(0)$, $\alpha_1$ also indicates ordering (heterocoordination) in the alloys.

Darken’s thermodynamic equation for chemical diffusion has been utilized for relating diffusion and $S_c(0)$. The computed values of the chemical diffusion coefficient exhibit positive deviations in the entire concentration range for the alloys. This further shows that the degree of chemical order is greater in Fe–Pt than in Ni–Pt. The effect of the atomic size mismatch variation on $S_c(0)$ in the long-wavelength limit is also related to the ordering phenomenon in the liquid binary alloys.

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