Concentration dependent structural parameters of liquid Al-Fe alloys

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Abstract. Square well potential is perturbed over Lebowitz solution of hard sphere mixtures to determine direct correlation function, $C^{0ij}(r)$ in repulsive and attractive regions under Mean Spherical Model Approximation [1]. Obtained direct correlation functions were employed to derive partial structure factors and then total structure factor, $S(k)$ in liquid Al-Fe alloy at different atomic percent of Al. Fourier transform of partial and total structure factors gives partial and total radial distribution functions, $g(r)$ from which partial and total coordination numbers and the partial nearest-neighbor distances were computed.

1. Introduction

Fe-Al alloys are widely used in the production of ductile cast iron, aluminized steel, and corrosion resistant materials, etc. [2, 3]. Due to its unique set of mechanical, magnetic and corrosion properties, the binary Fe-Al system also serve as the basis for the preparation of quasi crystalline and amorphous materials [4].

Since liquid state do not have systematic arrangements of particles, understanding the structure of liquids become all the more complicated and hence their solution and their applications become more important as the theory of symmetry cannot be applied nor the theory of probability in completeness. However, the structural properties of liquids can be explained on the basis of perturbation treatment on hard sphere reference system [5]. In this present work we have performed computations within the framework of square well (SW) model to evaluate partial and total structure factors and their derived associated properties like partial and total radial distribution functions, partial and total coordination numbers of Fe-Al alloys at various atomic fractions of Al.

2. Theory

We perturbed amended square well attractive part over the hard sphere reference system [6] within the Mean Spherical Model Approximation (MSMA) and so the Direct correlation function (DCF) can be written as while charge density and the potential are expanded as a Fourier series with wave vectors up to $G_{\text{max}}=12$ (a.u.)$^{-1}$. The different MT sphere radii ($R_{\text{MT}}$) used were 2.43 a.u., 2.9 a.u. and 2.43 a.u. for Cu, In and Te, respectively. The self-consistent potentials are calculated on a $17 \times 17 \times 17$ $k$-mesh in the Brillouin zone (BZ) and the convergence criterion is set to $10^{-4}$ Ry. The experimental lattice constant of $a=6.195$ Å and $c=12.418$ Å [24] are used to calculate the results presented here.
\[
C_{ij}(r) = \begin{cases} 
C_{ij}^{0}(r) & ; \quad 0 < r < \sigma_{ij} \\
- \varepsilon_{ij}/k_{B}T & ; \quad \sigma_{ij} < r < \lambda_{ij}\sigma_{ij} \\
0 & ; \quad r > \lambda_{ij}\sigma_{ij}
\end{cases}
\]  

(1)

Where, \( C_{ij}^{0}(r) \) is the DCF for a mixture of hard spheres obtained by Lebowitz for Percus -Yevick equation. Further, \( \sigma_{ii}, \varepsilon_{ii}, \) and \( \lambda_{ii} \) are the diameter, the potential energy depth and breadth respectively of the square well potential of the \( i^{th} \) species. The mixed parameters are determined by using Lorentz Berthelot rules [7]. The total correlation function \( h_{ij}(k) \) is related to the DCF through the generalized Ornstein-Zernikes’s equation for a system containing more than one species, which can be given as

\[
h_{ij}(k) = C_{ij}(k) + \sum_{1=1,2} \rho_{1} C_{11}(k) h_{j1}(k)
\]

(2)

The Ashcroft-Langreth partial structure factors \( S_{11}(k), S_{22}(k) \) and \( S_{12}(k) \) can be obtained by taking the Fourier Transformations of \( h_{ij}(k) \) given as

\[
S_{11}(k) = \left\{ 1 - \rho_{1} C_{11}(k) - \rho_{1}\rho_{2} C_{12}^{2}(k)/1 - \rho_{2} C_{22}(k) \right\}^{-1}
\]

(3)

\[
S_{22}(k) = \frac{[1 - \rho_{1} C_{11}(k)] S_{11}(k)}{1 - \rho_{2} C_{22}(k)}
\]

(4)

\[
S_{12}(k) = \frac{(\rho_{1}\rho_{2})^{1/2} C_{12}(k) S_{11}(k)}{1 - \rho_{2} C_{22}(k)}
\]

(5)

The total structure factors, \( S(k) \) in terms of partial structure factors, \( S_{ij}(k) \) and concentration is obtained as follows

\[
S(k) = \sum_{i=1}^{2} \sum_{j=1}^{2} (c_{i} c_{j})^{1/2} f_{i}(k) f_{j}(k) S_{ij}(k)
\]

(6)

Here \( f_{i}(k) \) and \( f_{j}(k) \) are the atomic scattering factors of \( i^{th} \) and \( j^{th} \) species respectively.

We obtain the partial radial distribution functions, \( g_{ij}(r) \) and also total radial distribution functions, \( g(r) \) by the Fourier inversion of the \( S_{ij}(k) \) and \( S(k) \) respectively. From \( g_{ij}(r) \) we compute the first coordination number \( \psi_{ij} \) through the following equation

\[
\psi_{ij} = 4\pi\rho_{ij} \int_{0}^{r_{\min}} g_{ij}(r) r^{2} dr
\]

(7)

Here \( r_{\min} \) is the first minimum of \( g_{ij}(r) \).
3. Results and discussion

The computations of the total structure factors were made using the pure parameters for Fe and Al as given in Table 1 at different concentrations of Al in Fe-Al alloys.

Table 1: Potential parameters of Fe-Al alloy

| Metals | $\sigma_{ii} (\text{Å})$ | $\epsilon_{ii}/K_B$ (K) | $\lambda_{ii}$ | $\rho_{ii}$ |
|--------|---------------------------|-------------------------|----------------|------------|
| Fe     | 2.28                      | 425.67                  | 1.76           | 0.0756     |
| Al     | 2.40                      | 64.72                   | 1.60           | 0.0646     |

It has been found that the computed total structure factors $S(k)$ and the corresponding pair-correlation functions $g(r)$ for liquid Fe-Al alloys at 50 K above the liquidus temperature shows a good agreement with the experimental results [8] at different concentrations of Al in Fe-Al alloys as given in Fig. 1 and Fig. 2. We found that as the atomic percent of Al increases, the peaks occur at the same position but the peak height of the total structure factor increases which is also shown in Fig.1. The peak positions and peak heights of the radial distribution functions of the alloys computed by Fourier transformation of total structure factors increase with increase in the concentration of Al in Fe-Al alloys as given in Table 2. Similar trend was observed experimentally by [8], there is a good agreement between our computed results and the experimental values.

Table 2: Details of structure factor and radial distribution function

| % of Al in Fe-Al alloy | Temperature (K) | Theoretical $S(k)$ | Experimental $S(k)$ | Theoretical $g(r)$ | Experimental $g(r)$ |
|------------------------|-----------------|--------------------|--------------------|--------------------|--------------------|
| 70                     | 1493            | 2.90               | 2.44               | 2.40               | 2.45               | 2.95               | 2.54               | 3.05               |
| 78                     | 1453            | 2.90               | 2.48               | 2.92               | 2.41               | 2.52               | 3.14               | 2.60               | 2.77               |
| 82                     | 1463            | 2.90               | 2.50               | 2.92               | 2.49               | 2.59               | 3.18               | 2.63               | 2.79               |
| 86                     | 1433            | 2.90               | 2.52               | 2.92               | 2.09               | 2.60               | 3.23               | 2.63               | 2.91               |

Table 3: Partial and total coordination numbers

| % of Al in Fe-Al alloy | $\psi_{11}$ | $\psi_{22}$ | $\psi_{12}$ | $\psi_{\text{total}}$ |
|-----------------------|-------------|-------------|-------------|-----------------------|
| 70                    | 7.961       | 6.125       | 0.018       | 14.104                |
| 78                    | 7.960       | 6.265       | 0.021       | 14.246                |
| 82                    | 7.945       | 6.563       | 0.022       | 14.531                |
| 86                    | 7.949       | 6.871       | 0.022       | 14.842                |

Table 4: The partial nearest-neighbour distances $r$ (in Å)

| % of Al in Fe-Al alloy | $r_{(\text{Fe-Fe})}$ | $r_{(\text{Al-Al})}$ | $r_{(\text{Fe-Al})}$ |
|-----------------------|-----------------------|-----------------------|-----------------------|
| 70                    | 2.49                  | 2.33                  | 2.57                  | 2.41                  | 2.53                  |
| 78                    | 2.62                  | 2.54                  | 2.59                  | 2.53                  | 2.63                  |
| 82                    | 2.63                  | 2.55                  | 2.61                  | 2.53                  | 2.64                  |
| 86                    | 2.54                  | 2.57                  | 2.60                  | 2.54                  | 2.65                  |

From partial radial distribution function we compute the coordination number of the alloys at different concentration of Al. As seen from Table 3, the total coordination number increases with
increase in the concentration of Al i.e., from 70 to 86 atomic percent of Al in Fe-Al alloys and the values changes from 14.10 to 14.84. The obtained results indicate a linear dependence of the $K$, $r$ and $\psi$ values on the concentration.

The partial nearest-neighbor distances, $r_{(i-j)}$, estimated as the position of the first peak in $g_{ij}(r)$ of the liquid Fe-Al alloys, at 50K above the liquidus temperature are listed in Table 4. It is found that $r_{(Al-Al)}$ has the shortest neighbor distances and $r_{(Fe-Fe)}$ has the longest neighbor distance which follows the same trend as the experimental result reported by [8].

**Figure 1.** Total structure factor $S(k)$ versus $k$ at different concentrations of Al in Fe-Al alloy; (→) theoretical values, (o o o) experimental values.
Figure 2. The pair correlation function g(r) versus r at different concentrations of Al in Fe-Al alloy: (→) theoretical values, (o o o) experimental values.

4. Conclusions
Structure factors and radial distributions of Fe-Al alloys at different atomic percent of Al were calculated at 50K above the liquidus temperature. Computed results were compared with the available experimental values and we found a good agreement between them. The coordination number increases with increase in the concentrations of Al. It was also found that the partial nearest-neighbor distances, \( r_{(i-j)} \), shows a reasonable dependence on the concentration of Al atomic percent and was observed to follow the same trend as the experimental results. Therefore, perturbation of Square well...
attractive tail over hard sphere reference system serves well for the study of static structure factor and derived properties of liquid binary alloys.

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