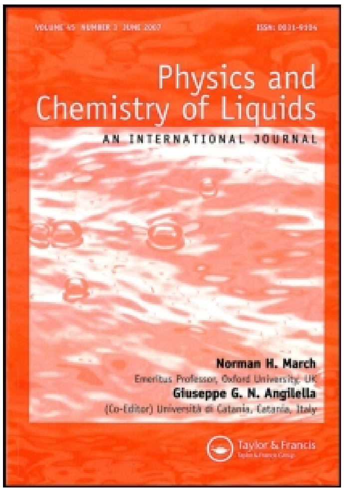

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Partial Coordination Numbers in Pb-Bi Molten Alloys

B. C. Anusionwu[a]
[a] Department of Physics, Federal University of Technology, Owerri, Nigeria

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PARTIAL COORDINATION NUMBERS IN Pb–Bi MOLTEN ALLOYS

B.C. ANUSIONWU

Department of Physics, Federal University of Technology, Owerri, Nigeria

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The study of the partial coordination numbers for the Pb–Bi liquid alloy showed an increase in the partial coordination number for the Pb–Pb species at 0.75 atomic fraction Bi. This anomaly suggests that the Pb atoms manifest some level of homocoordination within the neighbourhood of that concentration. It also provides an explanation for the observed change in the total coordination number of the liquid alloy at high Bi concentration.

Keywords: Metallic alloys; Coordination numbers; Liquid alloy

PACS: 61.25 Mv

1 INTRODUCTION

Recent research efforts in the theoretical study of liquid binary alloys are devoted to an understanding of compound formation in the systems incorporating the concept of formation of complexes and local ordering [1–3]. These studies are of increasing research interest due to the possibility of using them to predict the glass forming abilities of these molten alloys and in addition, the possible alloy concentrations where complexes exist [4]. Structural studies by Gopala Rao and Satpathy on Cu–Bi [5] and Gopala Rao and Das [6] have also been used to suggest liquid alloy concentrations where complex activities are expected.

Pb–Bi liquid alloy on its part has also aroused research interest and it has been studied previously by some authors [7–9]. These studies indicate that it is ‘anomalous’, partly because it manifests a small positive departure from linearity in the concentration dependence of liquid alloy volume. Further, Sharrah and Petz [8] observed a change in its total coordination number at high Bi concentrations. To explain the observed features of the alloy, Neumann *et al.* [9] suggested a possible structural arrangement of atoms with a preference of unlike atomic pairs.

To understand further the observed change in the coordination number of Pb–Bi alloy at high concentrations of Bi, it becomes worthwhile to determine in some detail the partial coordination numbers of the liquid alloy and to observe its trend with variation in Bi concentration.

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In this brief presentation, one proceeds by obtaining the partial pair distribution functions of the Pb–Bi liquid alloy at various concentrations of Bi from the partial structure factors obtained by [10] from the experimental data of [9]. The quantities obtained are then used to make some inferences about the liquid alloy.

The next section gives a brief outline of the theoretical background. Section 3 discusses the results of the calculations and conclusions are outlined in Section 4.

2 THEORETICAL FORMULATIONS

The Lebowitz's solution of the Percus–Yervick (PY) equation with a perturbation treatment can be applied to obtain the partial structure factors $S_{ij}(q)$ of a system of liquid binary alloys (where q is the atomic position in reciprocal space).

We define the direct correlation functions in the usual way appropriate for a hard sphere system with a wall [6,11]:

$$C_{ij}(r) = \begin{cases} C_{ij}^{hs}(r), & 0 \leq r \leq \sigma_{ij} \\ -\beta\varepsilon_{ij}, & \sigma_{ij} \leq r \leq A_{ij}\sigma_{ij} \\ 0, & r > A_{ij}\sigma_{ij} \end{cases} . \tag{1}$$

where $\beta = (1/K_B T)$ and C_{ij}^{hs} is the Percus–Yervick [11] solution for the hard spheres. σ_{ij}, ε_{ij}, and A_{ij}, represent the hard sphere diameter, depth and breadth of the square well respectively. i and j represent the ith and jth components of the binary alloy.

The Fourier transform $C_{ij}(q)$ of the direct correlation function $C_{ij}(r)$ has been taken from literature [12,13]. The partial structure factors are obtained from the combination of the $C_{11}(q)$, $C_{22}(q)$ and $C_{12}(q)$ [13] as

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

$$S_{22}(q) = \left[1 - \rho_{22}C_{22}(q) - \rho_{11}\rho_{22}C_{12}^2(q)/(1 - \rho_{11}C_{11}(q))\right]^{-1} \tag{3}$$

$$S_{12}(q) = \frac{(\rho_{11}\rho_{22})^{1/2}C_{12}(q)}{[(1 - \rho_{11}C_{11}(q))(1 - \rho_{22}C_{22}(q)) - \rho_{11}\rho_{22}C_{12}^2(q)]} \tag{4}$$

where 1 and 2 represent the first and second components of a binary alloy and ρ_{ii} represent the number density of the ith component. The total structure factor $S(q)$ is related to the partial structure factors by the relation

$$S(q) = \sum_{j=1}^{2}\sum_{i=1}^{2}(c_i c_j)^{1/2}\frac{f_i(q)f_j(q)}{c_1 f_1^2(q) + c_2 f_2^2(q)} S_{ij}(q) \tag{5}$$

$f_i(q)$ and $f_j(q)$ are the atomic scattering factors of the ith and jth component respectively. c_i and c_j are the atomic fractions of components i and j respectively.

The partial pair distribution functions $g_{ij}(r)$ are obtained from the partial structure factors $S_{ij}(q)$ by the relation

$$g_{ij}(r) = 1 + (2\pi^2 n_o r)^{-1}\int_0^\infty q(S_{ij}(q) - \delta_{ij})\sin(qr)dq \tag{6}$$

where $n_0 = (\rho_{ii}\rho_{jj})^{1/2}$. The partial coordination numbers Z_{ij} can now be obtained from the expression

$$Z_{ij} = 2\int_{r_o}^{r_m} 4\pi r^2 n_o g_{ij}(r)dr \tag{7}$$

where r_o denote the position for the beginning value of the partial pair distribution function and r_m is the position of the peak value.

3 DISCUSSION OF RESULTS

The partial pair distribution function for Pb–Bi melt at 773 K have been calculated from the partial structure factors obtained by [10]. The experimental data used by [10] in the determination of the partial structure factors is due to Neumann *et al.* [9].

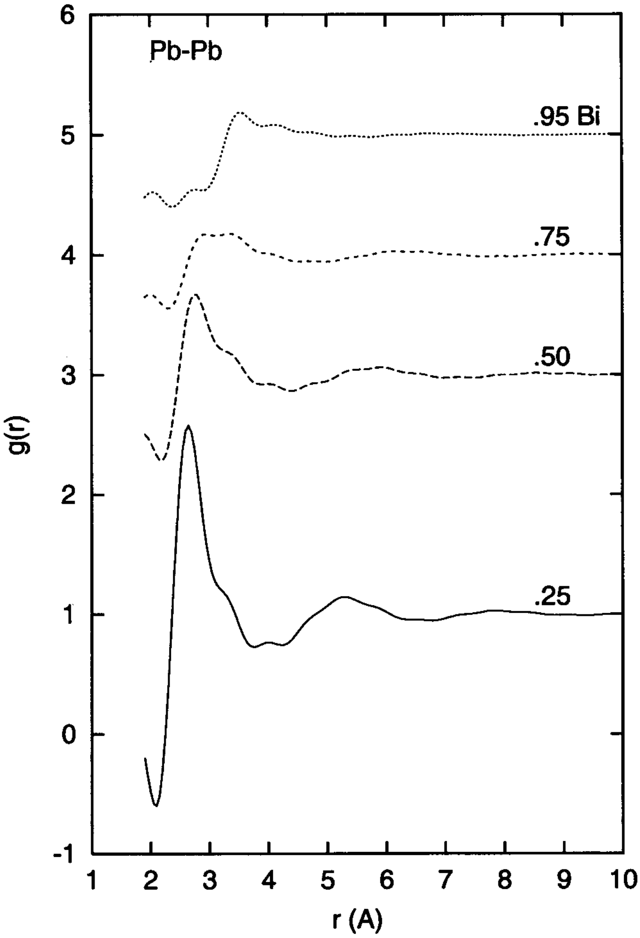

FIGURE 1 Partial pair distribution function $g_{\mathrm{Pb-Pb}}(r)$ *vs.* r(°A) for Pb–Bi molten alloys at various concentrations of Bi.

Figure 1 shows the partial pair distribution function $g_{\mathrm{Pb-Pb}}(r)$ for the Pb–Pb pair. The figure shows that the magnitude of the first peak for $g_{\mathrm{Pb-Pb}}(r)$ decreases with increase in the concentration of Bi. The hump to the right of the first peak at low concentration of Bi, increases to a twin peak at 0.75 atomic fraction Bi and finally as the principal peak at 0.95 atomic fraction of Bi. The presence of the twin peak signifies a possibility of some complex interactions among the atoms of the Pb specie.

Figure 2 shows the partial pair distribution function $g_{\mathrm{Bi-Bi}}(r)$ for the Bi–Bi pair. The principal peak increases with the concentration of Bi atoms. However, $g_{\mathrm{Pb-Bi}}(r)$ (Fig. 3) for the Pb–Bi pair exhibits a decrease in the principal peak with a simultaneous broadening of the peak as the concentration of Bi atom increases. The hump to the right of the peak develops to a subsidiary peak at higher concentrations of Bi.

The results of the calculation of the partial coordination numbers (Z_{ij}) of the Pb–Bi melt at the given concentrations 0.05, 0.25, 0.50, 0.75 and 0.95 atomic fractions of Bi appear revealing. One expects that for an alloy Pb–Bi exhibiting features of a uniform mixture [10], the partial coordination numbers ($Z_{\mathrm{Pb-Pb}}$) for the Pb–Pb pairs will

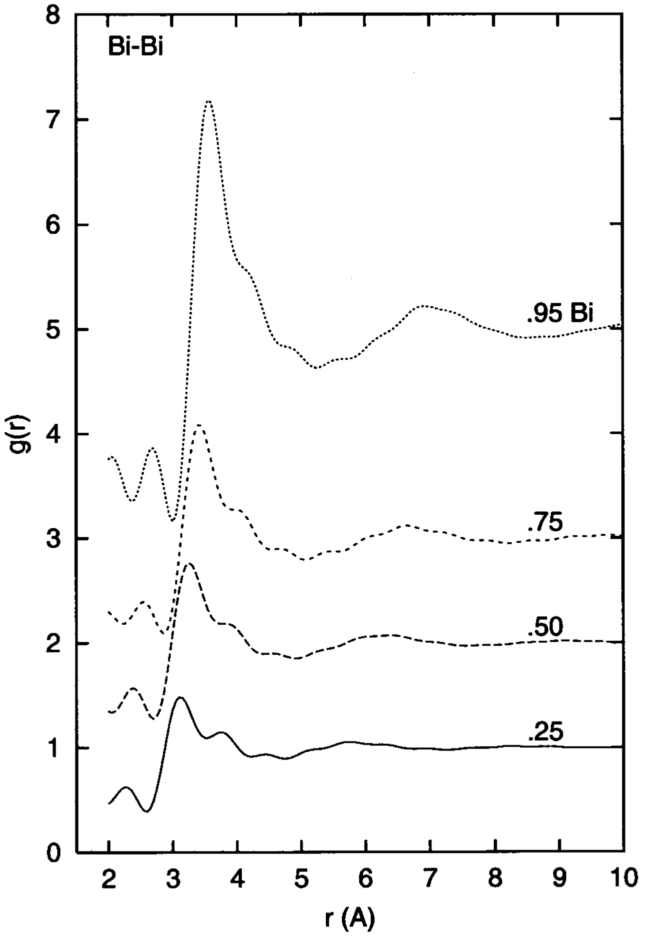

FIGURE 2 Partial pair distribution function $g_{\mathrm{Bi-Bi}}(r)$ vs r(°A) for Pb–Bi molten alloys at various concentrations of Bi.

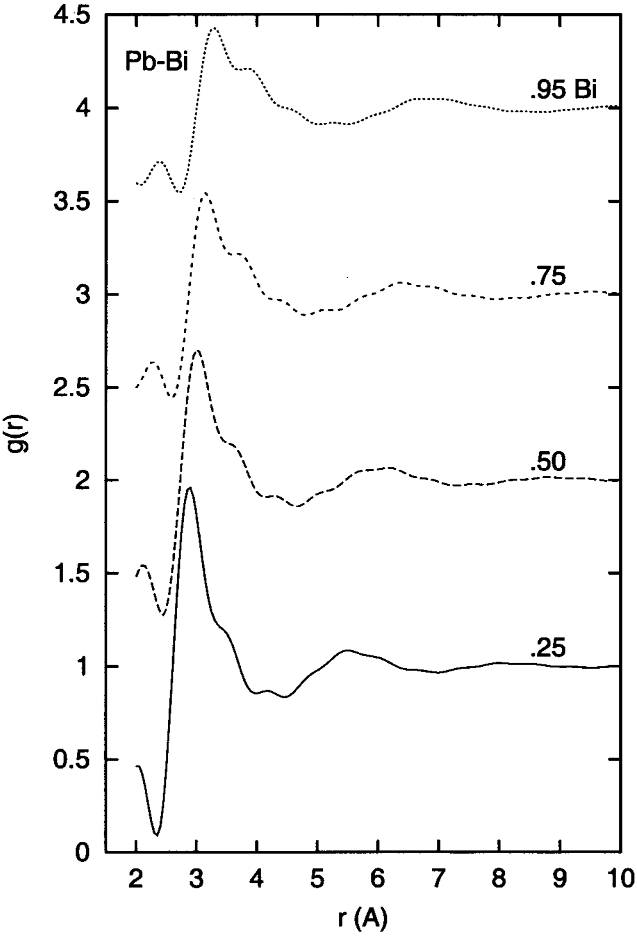

FIGURE 3 Partial pair distribution function $g_{\mathrm{Pb-Bi}}(r)$ *vs.* r(°A) for Pb–Bi molten alloys at various concentration of Bi.

decrease with increase in the concentration of the Bi atoms while the coordination number ($Z_{\mathrm{Bi-Bi}}$) for the Bi–Bi pairs will increase. The results in Fig. 4 and Table I show this trend, however, the coordination numbers ($Z_{\mathrm{Pb-Pb}}$) for the Pb–Pb configuration show an increase at about 0.75 atomic fraction of Bi. Note that, at this concentration, the partial pair distribution function for the Pb–Pb pair showed a twin peak. An increase in the partial coordination number at this concentration may be easily understood if we suggest the existence of a low level of segregation or homo-coordination of the Pb species around this concentration. Such segregation tendencies will undoubtedly lead to changes in manifest physical and structural properties of the liquid alloy within the affected region. In addition, where quenching is required, this same segregation property can affect the structural properties of the alloy in the solid state. Therefore, the observed change in the total coordination number of the liquid Pb–Bi alloy at high Bi concentration by Sharrah and Petz [8] will possibly be due to this segregation behavior manifested by lead atoms in this region.

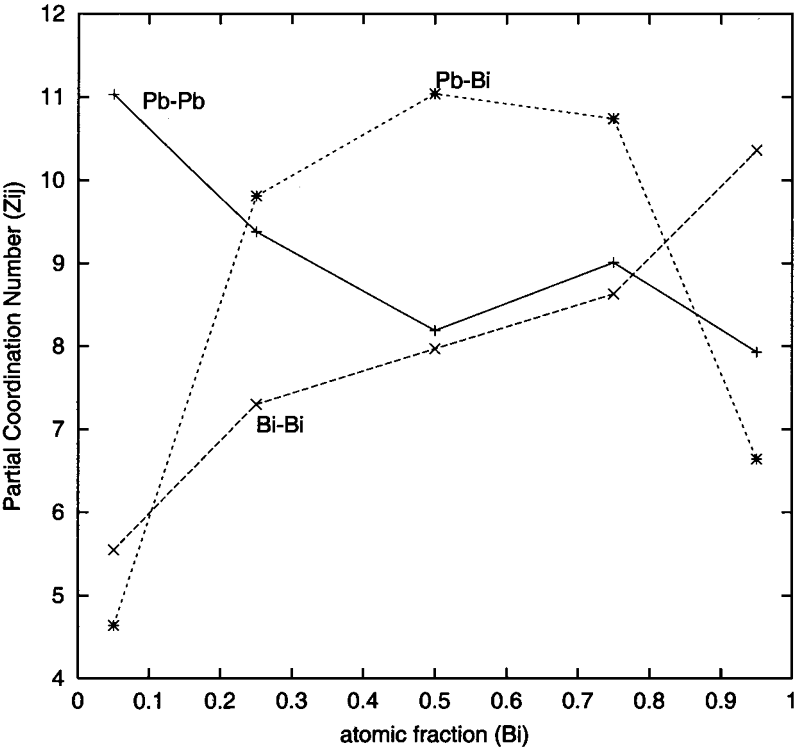

FIGURE 4 Partial coordination numbers Z_{ij} *vs.* concentration of Bi lines (Pb–Pb), dashes (Bi–Bi) and dots (Pb–Bi).

TABLE I Partial coordination numbers for Pb–Bi molten alloy

Concentration (Bi)	Z_{Pb-Pb}	Z_{Bi-Bi}	Z_{Pb-Bi}
0.05	11.30	5.55	4.64
0.25	9.38	7.30	9.38
0.50	8.19	7.97	11.04
0.75	9.01	8.63	10.74
0.95	7.93	10.36	6.64

The partial coordination number ($Z_{\mathrm{Bi-Bi}}$) for the Bi–Bi configuration increases with increase in Bi concentration. This is expected for an alloy of ideal mixing because at higher concentration of Bi, lesser Pb atoms will be present. The partial coordination numbers ($Z_{\mathrm{Pb-Bi}}$) for the Pb–Bi configuration have the maximum value at equiatomic concentration. This result is consistent with the results of Gopala Rao [6] for Mg–Bi molten alloy.

4 CONCLUSION

This study suggests that the manifest behaviour of the Pb–Bi liquid alloy at high Bi concentrations is as a result of the segregation tendencies of Pb atoms within this region.

The Bi atoms do not show any indication of segregation throughout the concentration range and hence have mixed evenly with the Pb clusters.

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