Molecular Dynamics Simulation of Amorphous Pd$_{80}$Si$_{20}$ Alloy

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Molecular dynamics (MD) simulation of an amorphous Pd$_{80}$Si$_{20}$ alloy has been carried out to know the microscopic structure in this system. After adjusting parameters of the proposed pair potentials, we have obtained similar structure factor of the simulated system to the experimental one and found some trigonal prismatic structures in the simulated system, which are not always predominant. Most probable coordination number of Pd around Si is 7, which is the same as that obtained by Distorted Prismatic Packing model proposed by Gaskell.

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I. Introduction

It is well known that the Pd–Si system makes amorphous alloy easily, and its properties have been well defined. As for its macroscopic and microscopic structures, they are studied both theoretically and experimentally. Gaskell proposed a structural model for metal-metalloid glasses and successfully reproduced a structure factor of amorphous Pd$_{80}$Si$_{20}$ alloy (a-Pd$_{80}$Si$_{20}$). In his model, regular, symmetric and trigonal prisms were constructed and then distorted to accommodate Si atoms there. It is interesting to know whether such kind of prisms are realized by molecular dynamics (MD) simulation, where pair potential form is just assumed. The MD simulation can give bulk properties of the system with a periodic boundary condition. Since choice of the potential form is not sensitive to the resulting structure factors, the agreement between experimental and computed structure factors does not always warrant validity of the potentials. Nevertheless, an agreement should be required as a first order check on the validity of models. The potential form for this system has not been revealed so far. However, since conduction electrons should screen ionic potentials, a pair potential of the Yukawa form and a short range repulsion is employed in this work tentatively.

\[ u_{ij}(r) = z_i z_j \exp\left(-\lambda r\right)/r + b \exp\left((r_0^{ij} - r)/\sigma\right) \]

where $z$, $\lambda$, $b$, $r_0^{ij}$ and $\sigma$ are considered to be the parameters. Although some chemical interaction exists between nearest neighbor Pd and Si, the MD simulation of this system may be carried out like SiO$_2$ and graphite, which were considered to be purely ionic systems.

II. Calculation and Results

The basic cell contains 410 Pd and 102 Si atoms. The side length of the basic cell, that is 1.96288 nm, is determined according to the density of this system. Initially, 512 regular lattice points are generated in the basic cell and then 102 points are randomly selected among them as the positions for Si. Then, The isothermal MD simulation proposed by Woodcock were carried out at first at 2000 K and subsequently at room temperature with a time increment of 2 fs. After annealing of the system, the structure factor of the simulated system is compared with the experimental one with an adjustment of the parameters $z$, $r_0^{ij}$, and $\sigma$ in the pair potential. This procedure is continued until satisfactory agreement is attained between them. After that, additional 1000 time steps
simulation was made to anneal the system fully and then the system is quenched to the room temperature (298 K), where diffractional experiments were carried out. At the room temperature, the radial distribution functions are accumulated over 600 time steps. Figure 1 shows a final result of the simulated structure factor along with the experimental ones(1)–(2)(7). The structure factor of the simulated system is calculated with the partial pair correlation functions and atomic scattering factors using the Debye equation.

\[
Q[S(Q) - 1] = \sum x_i x_j f_i(Q) f_j(Q) [\sum x_i f_i^2(Q)]^{-1} \times \int_0^{R_0} 4\pi \rho_0 [G_0(r) - 1] \sin (Qr) \, dr
\]

where \(x\) is the atomic fraction, \(f\) the atomic scattering factor(8) including anomalous dispersion terms(9) for Mo K\(\alpha\) X-ray radiation, \(\rho_0\) the average number density, and \(r^*\) half of the side length. The parameters of the pair potential are listed in Table 1.

### III. Discussion

The structure factors determined by MD and X-ray diffraction are Fourier transformed to obtain radial distribution functions \(G(r)\)'s, which are depicted in Fig. 2. Agreement between experimental and computed \(G(r)\)'s are satisfactory. Some characteristics of the simulated system are listed in Table 2 along with the experimental ones. The coordination numbers of Pd around Si and Pd are calculated by,

\[
n_0 = 4\pi \rho_0 \int_0^{R_0} r^2 G_0(r) \, dr
\]

where \(R_0\) is the position where \(G(r)\) crosses unity second time. In amorphous systems, it is difficult to say where the first nearest neighbor interaction is extinct, though the first nearest neighbor coordination number largely depends on the interaction distance, that is, the upper limit of the integration in eq. (3). According to Palinkás et al.(10), we employed \(R_0\) as an end of the first nearest neighbor interaction. All of the coordination numbers of Pd around Si listed in Table 2 are almost 7, while the coordination number of Pd around Pd in this work is significantly different from other two resear-
chers', which might have arisen from the different upper limit of the integration in eq. (3). Figure 3 shows a comparison between the present distribution of the nearest neighbor's number of Pd around Si within $R_0$ and Gaskell's results\(^1\). They seem to be very similar, though $P(8)$ is larger than $P(6)$ in this work, while $P(6)$ is larger than $P(8)$ in Gaskell's result. Figure 4 shows the stereoscopic views of the nearest neighbors of Pd around Si, which are arbitrarily selected from the instantaneous configurations of the system. One may see distorted prismatic configurations, (a) and (b) as well as non-prismatic configurations, (c) and (d). The fact that the predominant coordination number of Pd around Si is 7 leads us to a configuration similar to the one pointed out by Gaskell (Fig. 1(b) in Ref. (1) which is quite similar to Fig. 4(b) in this work). Although judgment of whether the configuration is (distorted) prismatic or not is not so clear, (distorted) prismatic configurations seem not to be predominant in our model. In this regard the structure of our model seems to be in a much relaxed state than that of Gaskell's. In spite of this fact, it sounds interesting that the primitive model presented here can at least reproduce, though not predominantly, the prismatic structure.

IV. Conclusion

The MD simulation of a-Pd$_{80}$Si$_{20}$ has been carried out with a primitive pair potential. The
reproduced the experimental one satisfactorily and some distorted prismatic structures can be realizes, though not predominantly. Possibility of the MD simulation for the elucidation of microscopic structure has been shown for the amorphous systems.

### REFERENCES


### Table 2 The characteristics of the radial distribution functions.

<table>
<thead>
<tr>
<th>Interaction</th>
<th>$r_i$/nm</th>
<th>$n_i$</th>
<th>Upper limit of integration/nm</th>
<th>Method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pd Si</td>
<td>0.234</td>
<td>7.1</td>
<td>0.262</td>
<td>MD</td>
</tr>
<tr>
<td></td>
<td>0.242</td>
<td>7.2</td>
<td>—</td>
<td>N</td>
</tr>
<tr>
<td></td>
<td>0.242</td>
<td>7.1</td>
<td>0.265</td>
<td>DPP</td>
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<tr>
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<td>0.308</td>
<td>MD</td>
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<tr>
<td></td>
<td>0.281</td>
<td>10.4</td>
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<td>N</td>
</tr>
<tr>
<td></td>
<td>0.283</td>
<td>10.5</td>
<td>—</td>
<td>DPP</td>
</tr>
</tbody>
</table>

$r_i$: probable nearest neighbor distance between $i$ and $j$, $n_i$: coordination number of $i$ around $j$, MD: molecular dynamics, N: neutron diffraction$^{(2)}$, DPP: distorted prismatic Packing model$^{(1)}$, and —: not specified in literature.