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分子動力学計算を用いた液体 Sn の拡散挙動解析

Time Series Analysis for Diffusion Behavior in Liquid Sn using Molecular Dynamics Simulation

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1. Introduction

Self-diffusion coefficients in liquid metal were measured under convection-free conditions, such as under microgravity[1)] or with stable density layering[2)] on the ground. On the other hand, it is useful to use Molecular Dynamics (MD) simulation to analyze the microscopic behavior of self-diffusion in liquid metal. It has been reported that liquid Sn has a unique structure called a shoulder in the pair distribution function, where is the probability of the existence of atoms increases at the slightly larger position than the position at the first peak, from scattering experiments[3, 4)]. However, it is not clear how the shoulder affects the microscopic behavior of self-diffusion in liquid Sn. The objective of this study is to clarify the effect of the shoulder in liquid Sn on the self-diffusion behavior using MD simulation.

2. Simulation method

MD simulation was conducted using LAMMPS package (version: 19 Sep 2019)[5)]. The modified embedded-atom method force field was used for the pair potential of Sn[6)]. 4096 atoms arranged in a cubic simulation box with standard periodic boundary conditions have been considered. Each atom is given a serial number (ID). The MD simulations were carried out by integrating Newton's equations of motion using velocity-Verlet algorithm with a time step of 1.0 fs. Before running the targeted calculation, the thermal equilibrium process was carried out to the simulation box according to the following procedure. First, the liquid sample was simulated at 2000 K during 3 ps using canonical ensemble (*NVT*) by means of a Nosé-Hoover thermostat to control temperature. Second, the simulation box was cooled down from 2000 to 573 K and equilibrated at 573 K during 18 ps using *NVT*. After the thermal equilibrium process, the targeted calculation was running during 100 ps using *NVT* and then time series data of atomic position was obtained.

3. Results

From the position data at starting the targeted calculation, the structure factor $S(Q)$ and the pair distribution function $g(r)$ were calculated. **Figure 1** shows the calculated structure factor. The calculated structure factor was in good agreement with the experimental data of liquid Sn[4)] from Q = 0 to 5 $Å^{-1}$ at 573 K. **Figure 2** shows the calculated pair distribution function, together with the typical local atomic configurations. As shown in **Fig. 2(a),** the next nearest neighbor atoms (yellow atoms in **Fig. 2(b)**) can be seen in the shoulder part of $g(r)$ from r = 3.5 to 5.1 Å. This implies the liquid Sn has characteristic local atomic configurations. Moreover, the mean-square diffusion depth (MSD) was calculated from the time series data of all position and self-diffusion coefficient of Sn in the simulation box was obtained. **Figure 3** shows the calculated self-diffusion coefficient of Sn. The value of self-diffusion coefficient of Sn at 100 ps was 2.29×10^{-9} m^2s^{-1} and that was in good agreement with the reference data under the microgravity condition at 573 K[1)].

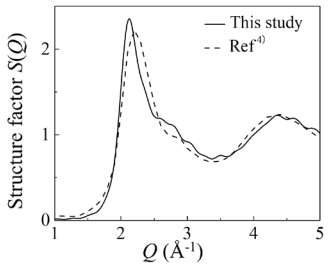

Figure 1. Calculated structure factor of liquid Sn at 573 K.

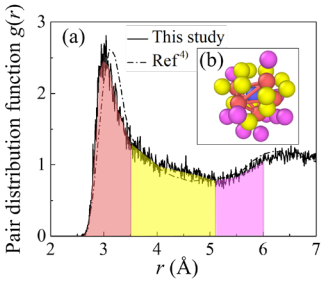

Figure 2. (a) Calculated pair distribution function of liquid Sn at 573 K and (b) visualization of typical local atomic configurations up to 6 Å. Each color area in (a) corresponds to the position of atoms from the center blue atom in (b). Red bars in (b) indicate the bonds between a pair of red atoms.

4. Discussion

To analyze the microscopic behavior of self-diffusion of Sn, some MSDs for individual Sn atoms were calculated. **Figure 4** shows the examples of the MSDs for the individual Sn atoms of ID450 and ID451 and for all atoms from 0 to 10 ps. The MSD for the ID450 atom was abnormally larger than that for the ID451 as seen in **Fig. 4**, although the constitute elements are only Sn. Therefore, the mechanism of such long-range diffusion should be discussed based on the atomic environments. Because of the presence of the next nearest neighbor atoms rather closer to the central atom as shown in **Fig. 2(b)**, the atomic motion is basically inhibited by the surrounded atoms. To understand the long-range diffusion, we need to consider some diffusion mechanisms such as a cooperative motion related to the next nearest neighbor atoms.

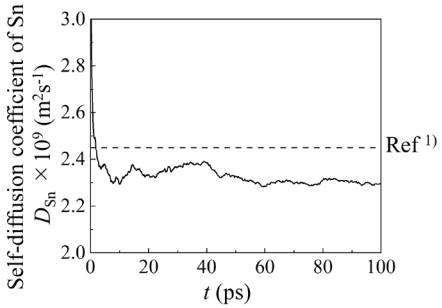

Figure 3. Calculated self-diffusion coefficient of liquid Sn at 573 K.

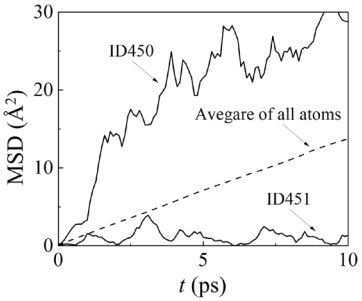

Figure 4. Examples of the mean-square diffusion depth of the individual Sn atoms.

5. Conclusion

The diffusion behavior in liquid Sn was analyzed using MD simulation. From the local atomic configurations, the next nearest neighbor atoms can be seen in the shoulder of the pair distribution function of Sn. The atomic motion of Sn is basically inhibited by the around atoms up to the next nearest neighbor. On the other hand, the long-range diffusion, which has abnormally larger MSD than the average MSD, was observed in spite of the above atomic environment.

Acknowledgement

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