JASMAC Conference of the Japan Society of Microgravity Application

P19

# シアーセル法と分子動力学計算による液体 Pb 中の自己および不純物拡散係数

# Self and impurity diffusion coefficient in liquid Pb measured by using shear cell technique and molecular dynamics

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## 1. Introduction

Diffusion coefficient in liquid metal is an important physical property for solidification simulation in casting. Molecular dynamics simulation (MD) is a useful method for simulating microscopic behavior of atom in liquid metal. It is necessary to obtain the diffusion coefficient experimentally as the benchmark for MD simulation. The objective of this study is to clarify how accurate impurity diffusion coefficient can be obtained in liquid Pb by comparing the diffusion coefficients obtained by using shear-cell technique and MD.

## 2. Experimental and analytical methods

The impurity diffusion coefficient of Sn in Pb was measured using the shear cell technique. Diffusion couples of pure Pb and Pb-5 at.%Sn alloy were placed in four capillaries (labeled Cap. A, B, C and D). Each sample was set in each capillary, as shown in **Fig. 1(a)**, so as to provide the stable density layering[1)]. Diffusion experiments were performed at 773 K for a diffusion time of 18000 s. After the diffusion time, each capillary was separated into 20 cells (labeled No.1-20). Each solidified sample was dissolved in a mixed acid solution and then the Sn concentration in the solution was analyzed by ICP-OES.

The MD simulation of self-diffusion of Pb and impurity diffusion of Sn in Pb-5 at.%Sn were performed. 13500 Pb atoms were arranged in a cubic simulation box which had an fcc structure. 675 atoms were replaced with Sn atoms only in impurity diffusion simulation. Calculations were carried out with PFP[2)] version 4.0.0 on Matlantis[3)]. Simulations of self-diffusion and impurity diffusion were performed under the NVT ensemble at 773 K with the time of 1 fs × 5000 steps and 10000 steps, respectively, after the equilibration with the time of 1 fs × 2000 steps. The simulations of impurity diffusion were performed four times under the same conditions. Diffusion coefficients were calculated from the mean square displacement in MD results. In the simulations of self-diffusion, the number density was 27.2 $nm^{-3}$. The number density for the simulation of impurity diffusion was obtained by linearly interpolating the number density[4)] with the concentration of Sn.

## 3. Results

**Figure 1(b)** shows the Sn concentration profiles obtained by using the shear cell technique. Floating objects appeared when the samples are dissolved in a mixed acid. The self-diffusion coefficient of Pb was 3.02

× $10^{-9}$ $m^2s^{-1}$ when number density was 27.2 $nm^{-3}$. The diffusion coefficients calculated by using MD were (3.61 ± 0.09) × $10^{-9}$ $m^2s^{-1}$ for Sn and (3.09 ± 0.02) × $10^{-9}$ $m^2s^{-1}$ for Pb.

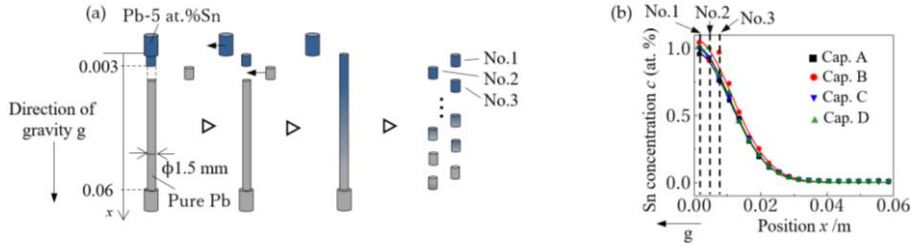


**Figure 1.** Shear cell technique and experimental results. (a) Schematics of the experimental procedure. (b) Sn concentration profiles for four capillaries. Each curve was calculated by the thick layer solution.

## 4. Discussion

According to **Fig. 1(b)**, some plots in Cap. B deviated from the thick layer solution. It is considered that the measured concentration dropped due to the floating object. The values of No.1 and No.2 were considered to be outliers. When the fitting was performed with excluding cells No. 1 and No. 2, the diffusion coefficient in Cap. B was corrected to be 3.02×$10^{-9}$ $m^2s^{-1}$. The obtained diffusion coefficient was (3.16±0.03)×$10^{-9}$ $m^2s^{-1}$ after the correction. The self-diffusion coefficient of Pb calculated by using MD increased with decreasing number density. The diffusion coefficient calculated by using MD was 13.9 % larger than that of shear cell technique. The density[4] in MD (9.26 × $10^3$ $kgm^{-3}$) was smaller than the actual density (1.044 × $10^4$ $kgm^{-3}$)[5]. Therefore, the calculated diffusion coefficient become larger with decreasing the number density.

## 5. Conclusion

By eliminating plots on concentration profile which dropped due to the floating object, the relative standard deviation of the obtained diffusion coefficient decreased. Diffusion coefficient calculated by using MD increased with decreasing number density.

## Acknowledgement

This study was supported by financial support from ENEOS Corporation, Grant-in-Aid for Kimura Foundry Co.,Ltd., Grant-in-Aid for Scientific Research(C) Grant Number JP23K04364, Grant-in-Aid for JSPS Research Grant Number JP20J14950, and Grant-in-Aid for JST SPRING, Grant Number JPMJSP2128.

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