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シアーセル法を用いた液体 **Pb** の自己拡散係数測定における安定密度配置の検証

Stable Density Layering for Self-Diffusion Measurement in Liquid Pb with a Shear Cell Technique

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1. Introduction

Self-diffusion coefficients of liquid metal are important to understand metallurgical phenomena. The self-diffusion coefficients of isotopes can be measured by changing the mass number of isotopes to be added to the diffusion samples. However, it is not clear the necessary density difference to achieve a stable density layering[1] which suppresses natural convection on the ground during the diffusion experiments. The objective of this study is to predict the boundary density difference for a stable density layering through the measurement of self-diffusion coefficients.

2. Experimental Procedure

Figure 1 shows a schematic illustration of the diffusion experiment using the shear cell technique[1] with four capillaries. To refer to impurity diffusion coefficients of Ag[2], the samples of pure Pb and PbAg alloy were used. In capillaries, alloy samples of PbAg containing isotopes of ^{204}Pb, ^{206}Pb, ^{207}Pb and ^{208}Pb were set on the bottom in the capillaries, which are named Caps. 204, 206, 207 and 208, respectively, while the samples of pure Pb were set on the top. The initial concentration of Ag in each PbAg alloy was 2 at.%. For the setting, the density was calculated under approximation of linear relationship between density and concentration of isotopes and reference of PbAg density[3]. The density of the lower side was estimated to be 5.0 kg/m^3 higher than the upper side in Caps. 207 and 208, while 17.2 kg/m^3 lower in Caps. 204 and 206. After evacuating the chamber containing the shear cell, the furnace was heated up to the diffusion temperature at 773 K. After homogenization, the diffusion process started. At the end of the diffusion time of 21600 s, the samples were divided into small cells mechanically and cooled down. The concentration of isotopes of Pb and Ag in each cell was analyzed by using ICP-MS and ICP-OES, respectively.
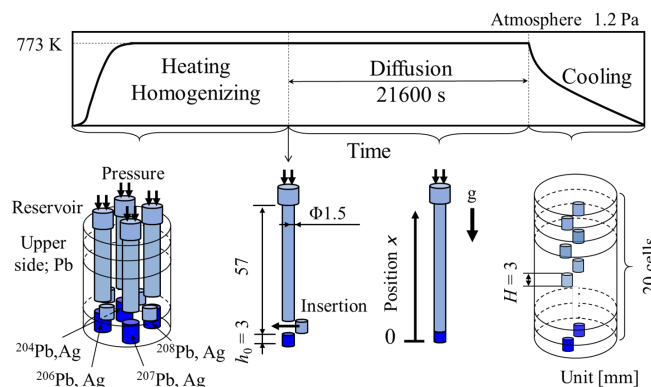

Fig.1 Schematic illustration of the diffusion experiment using the shear cell technique.

3. Results

Figure 2 shows the concentration profiles of isotopes [204]Pb and [208]Pb as the representative diffusion measurements. Concentration profiles of isotopes and Ag were fitted with the thick layer solution. The values of the self-diffusion coefficients D_{207Pb} and D_{208Pb} (3.05 × 10⁻⁹ and 2.88 × 10⁻⁹ m²/s) were in good agreement with the μg - reference data[4] within the error of 4.6 %. On the other hand, the values of self-diffusion coefficients D_{204Pb} and D_{206Pb} (8.50 × 10⁻⁹ and 9.70 × 10⁻⁹ m²/s) were about three times larger than the μg - reference data[4]. For referring to the impurity diffusion coefficients D_{Ag}, the values of D_{Ag} of Caps. 207 and 208 (3.53 × 10⁻⁹ and 3.53 × 10⁻⁹ m²/s) were in good agreement with the reference data[2] within the error of 2.8 %, while the values of D_{Ag} of Caps. 204 and 206 (5.85 × 10⁻⁹ and 5.99 × 10⁻⁹ m²/s) were about 1.6 times larger than the reference data[2].
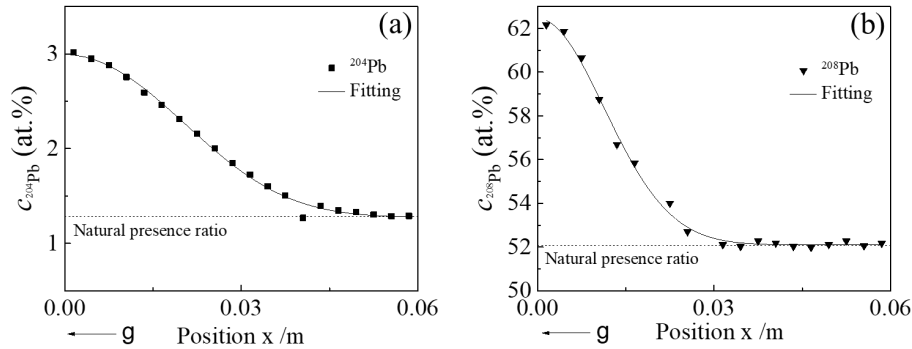

Fig.2 Concentration profiles at 773 K: (a) [204]Pb and (b) [208]Pb. The symbol "←g" indicates the direction of gravity.

4. Discussion

The density distribution of each capillary after the diffusion experiment was calculated from the concentration profiles of the isotopes and Ag under the approximation of the density in the same way as the initial density calculation. As a result, it was revealed that the density increases monotonically in the direction of gravity even at the end of diffusion for Caps. 207 and 208, while the density decreases monotonically for Caps. 204 and 206. Therefore, D_{207Pb} and D_{208Pb} could be measured with suppressing natural convection, where the density distributions were always larger density in the direction of gravity during the diffusion experiments.

5. Conclusion

Natural convection was suppressed in self-diffusion experiments with [207]Pb and [208]Pb, while was occurred in those with [204]Pb and [206]Pb. Therefore, the boundary density difference for a stable density layering exists between Caps. 207 and 206 due to the difference in initial density from pure Pb. The stability of the density layering can be predicted under approximation of linear relationship between density and concentration of isotopes.

References

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