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液体 Al 中における Sn の不純物拡散係数測定

Measurement of Impurity Diffusion Coefficient of Sn in Liquid Al

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1. Introduction

The impurity diffusion coefficient in liquid metals is an important physical property for understanding the crystal growth and the solidification process. Among liquid metals, liquid Al has a simple diameter whose pair distribution function can be represented by hard-sphere model. However, in the case of liquid Al, it is difficult to obtain accurate diffusion coefficients due to the gravitational segregation and effects of reaction with the graphite equipment. Another problem is that the thermodynamic interaction between solvent and solute is large. The objective of this study is investigating the effect of gravitational segregation, reaction with the graphite, and thermodynamic interaction on the impurity diffusion coefficient of Sn in Al.

2. Experimental Procedure

Diffusion experiments were conducted using the shear cell technique which was designed for the Russian satellite Foton missions[1)]. The raw materials of Sn and Al were weighed and cast in a graphite crucible so that the composition should be $Al_{97}Sn_3$ (named the cast sample of AlSn). The cast sample of AlSn was divided into 4 pieces in the order from the top in the direction of gravity at the fabrication stage, and they were labeled Capillaries A, B, C, and D from the top. Diffusion couples of pure Al and AlSn alloys were placed in four capillaries with ϕ1.5×60 mm. The AlSn alloys were placed at the bottom to provide the stable density layering with suppressing natural convection. The chamber containing the shear cell device which is made of graphite was evacuated, and the separated samples were heated to a diffusion temperature of 973 K. After homogenization, the intermediate cell was inserted and diffusion was started simultaneously in four capillaries. After 21600 s of diffusion time, each capillary sample was mechanically separated into 20 cells and cooled. Each removed sample was dissolved in mixed acid, and the solute concentration was analyzed using ICP-OES.

3. Results

Figure 1 shows the concentration profiles of Sn in liquid Al at 973 K. Due to gravitational segregation, the initial concentration of Sn (c_0) of Capillary A, which was located at the top of the gravity direction during the preparation of the cast sample of AlSn, was the lowest among all capillaries. On the other hand, c_0 of Capillary D was the highest among all capillaries, which was located at the bottom. Due to the reaction between the Al and the graphite cell, the first samples ($x_1 = 1.5 \times 10^{-3}$ m) of the concentration profiles were smaller than the second samples ($x_2 = 4.5 \times 10^{-3}$ m) because the samples were scrapped off when they were removed from the cell. Therefore, the impurity diffusion coefficients of Sn in Al (D_{SnAl}) were obtained by fitting thick layer solution to all plots except for plot of x_1. Since the concentrations of all samples in Capillary A were below the quantification of ICP-OES limit, diffusion coefficients were calculated only for Capillaries B, C and D. The coefficient of determination R^2 for all capillaries were greater than 0.995, indicating good fitting. From the fitting results, D_{SnAl} were 5.42×10^{-9} m^{2}s^{-1} for Capillary B, 6.27×10^{-9} m^{2}s^{-1} for Capillary C, and 6.57×10^{-9} m^{2}s^{-1} for Capillary

D. As a result, it was found that the capillary B, which was the highest in the downward direction of gravity, had the smallest D_{SnAl}, except for Capillary A, which was at the top of the cast sample of AlSn. On the other hand, Capillary D, which was from the lowest position, had the largest D_{SnAl}.

4. Discussion

Figure 2 shows the relationship between c_0 and D_{SnAl} and the relationship between c_0 and thermodynamic factor Φ_{SnAl}. The value of Φ_{SnAl} was calculated using the excess enthalpy corresponding to the composition of AlSn alloy[2)] and it indicates the thermodynamic interaction between the solute Sn and the solvent Al at c_0 of each capillary. The D_{SnAl} increased with increasing c_0. Although the tendency of Φ_{SnAl} indicates the concentration dependence of D_{SnAl}, the behavior of Φ_{SnAl} and the D_{SnAl} for the c_0 was opposite. Therefore, the experimental values were found to increase due to random errors independent of the concentration dependence. This random error was probably due to the fact that the first cell could not be acquired and used for fitting due to its reaction with the graphite cell.

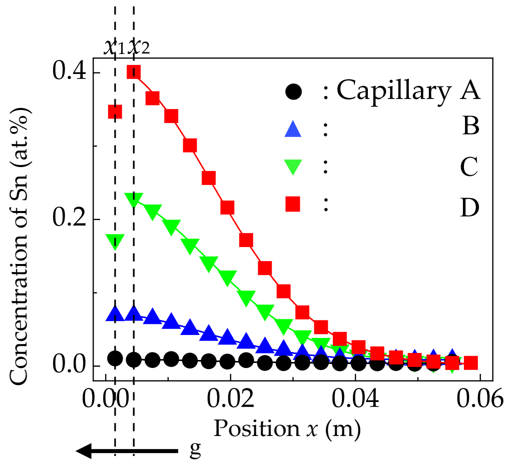

Fig. 1 Concentration profiles of Sn in liquid Al at 973 K. The symbol "←g" indicates the direction of gravity.

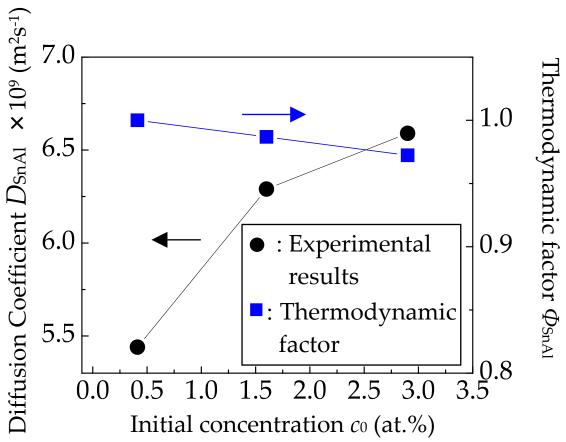

Fig. 2 Relationship between c_0 and D_{SnAl} and relationship between c_0 and thermodynamic factor Φ_{SnAl}.

5. Conclusion

The impurity diffusion coefficients of Sn in liquid Al D_{SnAl} were measured using the shear cell technique and stable density layering. The initial concentrations c_0 of the four capillaries were found be different due to gravitational segregation. The first sample could not be taken out due to the reaction with the graphite cell, so the concentration profiles were fitted by excluding the first sample. Although the D_{SnAl} was increased with increasing c_0, Φ_{SnAl}, which indicates the concentration dependence of D_{SnAl}, was decreased with increasing c_0. This is opposite behavior between Φ_{SnAl} and D_{SnAl}. Therefore, it can be considered to be due to a random error rather than concentration dependence.

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