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液体 Sn 中における Al および Au の不純物拡散係数測定

Measurement of Impurity Diffusion Coefficient of Al and Au in Liquid Sn

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1. Introduction

Impurity diffusion coefficient in liquid metals is an essential thermophysical property to understand the crystal growth and solidification of metals, so it is required to construct accurate prediction equations. Yamada *et al.*¹⁾ reported a prediction equation for the impurity diffusion coefficient D_{is} in liquid Sn near the melting point as shown in Eq. (1)

$$D_{is} = D_s^* \left(\frac{r_s}{r_i} \right) \Phi_{is} \quad (1)$$

D_s^* is the self-diffusion coefficient of the solvent, r is the atomic radius, and Φ_{is} is the thermodynamic factor. The subscripts i and s denote the solute and solvent elements, respectively. When the atomic radius ratio r_s/r_i is around 1 or smaller than 1, Eq. (1) reproduces the experimental values measured by our group¹⁾ within 10%. However, no experiments have been conducted using the shear cell technique²⁾ as the conditions where r_i is kept constant and the atomic weight of solute M_i is changed. Therefore, our group selected Al and Au as solute elements, the r_i of which is similar to Ag used in a previous experiment¹⁾. M_i of Al and Au are smaller and larger than M_{Ag} , respectively. The objective of this study was to investigate the effect of atomic weight on the impurity diffusion coefficient.

2. Experimental Procedure

Diffusion experiments were conducted using the shear cell technique²⁾ which was designed for the Russian satellite Foton missions²⁾. The initial concentration c_0 of Al in the SnAl alloy and Au in the SnAu alloy were set to 2 and 3 at.%, respectively. To achieve the stable density layering for suppressing natural convection, the samples of SnAl alloys and SnAu alloys were placed on the upper side and the lower side in the shear cell device, respectively. After evacuating the chamber containing the shear cell device, the furnace was kept at the diffusion temperature of 573 K while the samples were separated. After homogenization, the intermediate cell was inserted and the diffusion in each capillary was started simultaneously. At the end of the diffusion, each capillary sample was divided into 20 cells mechanically and cooled down. Each cell sample was dissolved in mixed acid and the solute concentration was analyzed by ICP-OES.

3. Results

Figure 1 shows the concentration profiles of experiments using Sn-2 at.%Al and Sn-3 at.%Au at 573 K. Each curve was obtained by fitting thick layer solution to 20 plots in each capillary. In Capillary B in **Fig. 1 (a)**, the plots at $x = 43.5$ and 49.5 mm, which were far above the lower limit of quantification in ICP-OES, were removed from the fitting as outliers. A small amount of Al was detected in the pure Sn sample used in experiments, so the concentration of Al c_{Al} was added to the equation of the thick layer solution. The coefficient of determination R^2 was larger than 0.995 in all experiments. The average of four measured values of D_{AlSn} and D_{AuSn} were 3.70×10^{-9} and 2.71×10^{-9} m²s⁻¹, respectively. The relative standard deviation of D_{AlSn} and D_{AuSn} were 3.83 and 0.53%, respectively.

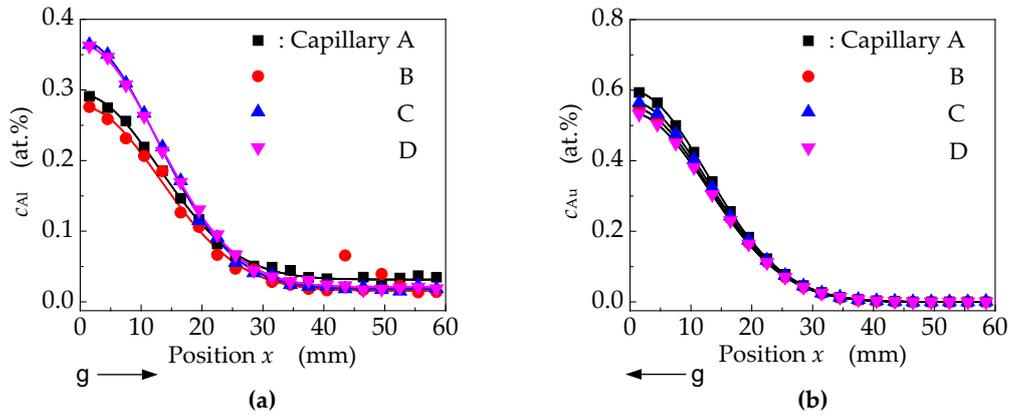


Fig. 1 Concentration profiles of solutes in liquid Sn at 573 K: (a) Sn-2at.%Al and (b) Sn-3at.%Au. The symbol “g→” indicates the direction of gravity.

4. Discussion

Figure 2 shows the relationship between atomic weight ratio M_{Sn}/M_i and impurity diffusion coefficient in liquid Sn D_{iSn} at 573 K. Since the error bars of Al and Ag did not overlap each other, there was a dominant difference between them due to the atomic weight and D_{AlSn} was larger than D_{AgSn} . Similarly, the error bars of Au and Ag did not overlap each other, so there was a dominant difference between them and D_{AuSn} was smaller than D_{AgSn} . Therefore, when comparing elements with the same r_i , D_{is} tends to become larger and smaller as M_i becomes smaller and larger, respectively.

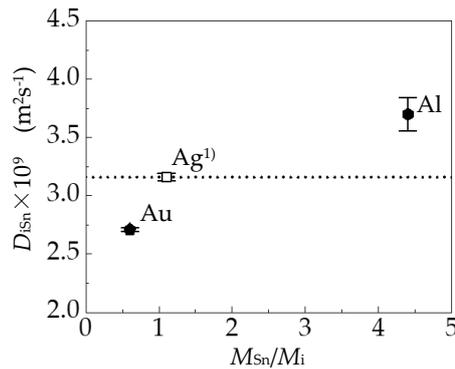


Fig. 2 Relationship between atomic weight ratio M_{Sn}/M_i and impurity diffusion coefficient in liquid Sn D_{iSn} at 573 K. The error bars indicate the standard deviation.

5. Conclusion

The effect of atomic weight on the impurity diffusion coefficient was investigated using Al and Au which have similar r_i to Ag at 573 K. The impurity diffusion coefficient in liquid Sn D_{iSn} becomes larger with a smaller atomic weight ($D_{AlSn}=3.70 \times 10^{-9} m^2 s^{-1} > D_{AgSn}$), while smaller with a larger one ($D_{AuSn}=2.71 \times 10^{-9} m^2 s^{-1} < D_{AgSn}$).

Acknowledgement

This study was supported by financial support by Kimura Foundry Co. Ltd, Grant-in-Aid for Scientific Research(C) Grant Number JP19K04990 and Grant-in-Aid for JSPS Research Grant Number JP20J14950.

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