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シアーセル法を用いた液体 Al 中の不純物拡散係数測定 Impurity diffusion measurement in liquid Al using shear cell technique

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1. Introduction

The diffusion coefficient in liquid metals is an important physical property for understanding the crystal growth and the solidification process. Natural convection reduces the accuracy of the measured diffusion coefficient. When the maximum density gradient is greater than 0, natural convection is suppressed in liquid Pb¹⁾. In this study, the diffusion experiments of liquid Al were performed, where the reaction of Al with the container material tends to occur²⁾. The objective of this study is to investigate the optimum experimental condition to suppress natural convection and the reaction with the container material.

2. Experimental Procedure

Diffusion experiments were conducted using the shear cell technique³⁾. Diffusion couples of pure Al and AlSn alloy were placed in four capillaries (labeled Cap. A, B, C, and D) made of graphite with $\phi 1.5 \times 60$ mm. The AlSn alloy, which has a higher density than pure Al, was set on the bottom side to realize stable density layering. Diffusion experiments were performed at 973 K with a diffusion time of 21600 s. After the diffusion measurements, the Sn concentration of the samples was analyzed using ICP-OES.

3. Results

Figure 1 shows the linearized representations of the obtained Sn concentration profile. The profiles can be approximated using a Gaussian function,

$$\ln(c(x, t) - c_1) = -\left(\frac{1}{2\langle X_{meas}^2 \rangle}\right)x^2 + \ln\left(\frac{(c_0 - c_1)h}{\sqrt{\pi\langle X_{meas}^2 \rangle/2}}\right) \quad (1)$$

$$\langle X_{meas}^2 \rangle = 2Dt + \langle X_{add}^2 \rangle \quad (2)$$

where x is the distance from the bottom of the samples; c_1 is the initial Sn concentration contained pure Al sample; t is the diffusion time; h is the thickness of the alloy sample; $\langle X_{meas}^2 \rangle$ is the measured mean-square diffusion depth; $\langle X_{add}^2 \rangle$ is additional mean-square diffusion depth, which was treated as $9.5 \times 10^{-7} \text{ m}^2$ ³⁾. Initial concentrations (c_0) in the AlSn alloys and $\langle X_{meas}^2 \rangle$ were calculated by fitting Eq. (1) to the concentration profile above the lower limit of ICP-OES quantification. The sample in the nearest cell from the bottom ($x = 1.5 \times 10^{-3} \text{ m}$) were excluded from the fitting concentration distributions because of sample loss due to the reaction with the graphite. The sample loss was larger when c_0 was higher. Then diffusion coefficients (D_{SnAl}) were calculated by Eq. (2). From the fitting results, D_{SnAl} and c_0 were $16.9 \times 10^{-9} \text{ m}^2\text{s}^{-1}$ and 0.05 at. % (Cap. A),

$13.3 \times 10^{-9} \text{ m}^2\text{s}^{-1}$ and 0.46 at. % (Cap. B), $6.93 \times 10^{-9} \text{ m}^2\text{s}^{-1}$ and 1.55 at. % (Cap. C) and $6.53 \times 10^{-9} \text{ m}^2\text{s}^{-1}$ and 2.89 at. % (Cap. D), respectively.

4. Discussion

According to **Fig. 1**, the coefficients of determination (R^2) of the fitted lines in Cap. C and Cap. D were larger than 0.99. In contrast, the plots of Cap. A and Cap. B were scattered from the fitted lines with R^2 values of 0.47 and 0.86, respectively, which indicates the disturbance of natural convection.

Figure 2 shows the relationship between the diffusion coefficient and the maximum density gradient $|\text{d}\rho/\text{d}x|_{\text{max}}$ on the horizontal axis. The $|\text{d}\rho/\text{d}x|_{\text{max}}$ was calculated from the density distribution which was approximated from concentration distributions after the diffusion based on the density data⁴⁻⁷. When $|\text{d}\rho/\text{d}x|_{\text{max}}$ was small, the measured D_{SnAl} decreased with increasing $|\text{d}\rho/\text{d}x|_{\text{max}}$. At least over $2.0 \times 10^4 \text{ kg/m}^4$, D_{SnAl} became constant at $6.7 \times 10^{-9} \text{ m}^2\text{s}^{-1}$. This means when the maximum density gradient is sufficiently large, natural convection is suppressed in the capillary, and thus their constant D_{SnAl} values seem to be the exact diffusion coefficient.

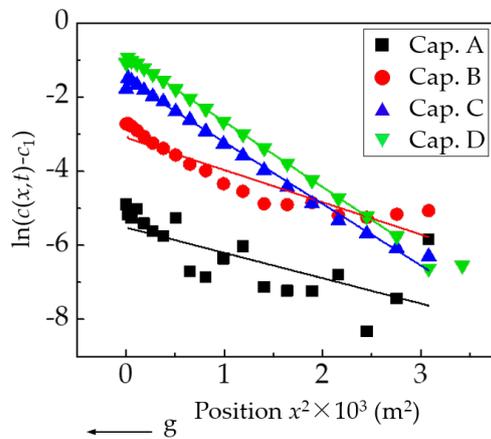


Figure 1. Concentration profiles of Sn in liquid Al at 973 K. The symbol “←g” indicates the direction of gravity.

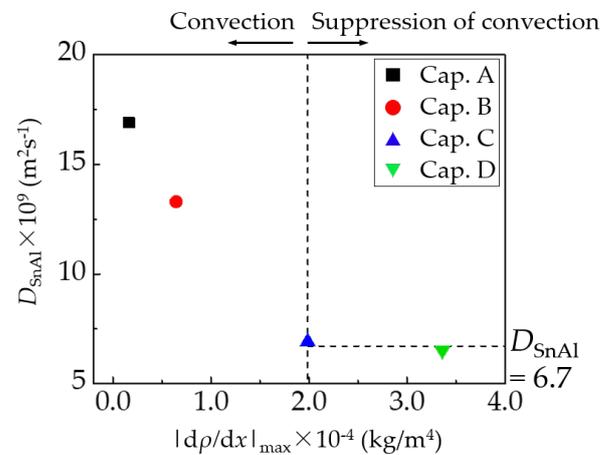


Figure 2. Relationship between measured D_{SnAl} and the maximum density gradient

5. Conclusion

When the maximum density gradient is larger than $2.0 \times 10^4 \text{ kg/m}^4$, natural convection is suppressed. Under the above condition, the measured diffusion coefficient is considered constant. To suppress natural convection and reduce sample loss, c_0 should be set in the range from 1.55 to 2.89 at. %.

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References

- 1) M. Shiinoki, Y. Nishimura, K. Noboribayashi, and S. Suzuki: Metall. Mater. Trans. B., **52B** (2021) 3846
- 2) A. Yamanaka, K. Noboribayashi, M. Shiinoki, and S. Suzuki: JASMAC-33 abstract (2021) P-11.
- 3) S. Suzuki, K. H. Kraatz, and G. Froberg: Ann. NY. Acad. Sci., **1027** (2004) 169.
- 4) K. Bornemann, and F.Sauerwald: Z. Metallkde., **14** (1922) 155.
- 5) A. Knappwost, and H. Restle: Z. Elektrotech., **58** (1954) 112.
- 6) F. Sauerwald, and K. Töpler: Z. Anorg. Chem., **157** (1926)130.
- 7) G. Doege: Z. Naturf., **21a** (1966) 266.

