JASMAC Conference of the Japan Society of Microgravity Appllication

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安定密度配置とシアーセル法を用いて測定した液体 Sn 中における Cu の不純物拡散係数

Impurity Diffusion Coefficient of Cu in Liquid Sn measured using the Stable Density Layering and the Shear Cell Technique

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1. Introduction

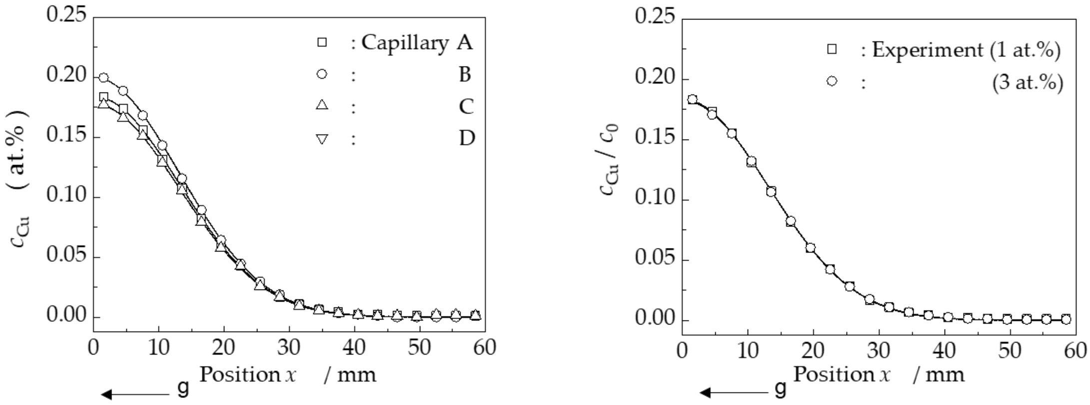
Impurity diffusion coefficients in liquid metals are important values to understand the mechanism of solidification and crystal growth. Yamada *et al*.[1)] reported that impurity diffusion coeffcients D_{is} in liquid Sn at 573 K can be expressed to be proportional to the product of the ratio of the atomic radii r_s/r_i (s: solvent, i: impurity) and thermodynamic factor Φ_{is}. Here, the self-diffusion coefficient D^*_s is the proportional constant. This equation is demonstrated to be valid when r_s/r_i is around 1 or smaller than 1. Our group[2)] measured the impurity diffusion coefficient of Cu in liquid Sn D_{CuSn} at 573 K as a condition that r_s/r_i is enough larger than 1. As a result, D_{CuSn} was measured to be 16 % smaller than estimated value calculated from the equation reported by Yamada *et al*.[1)]. There is a possibility that the samples of the SnCu alloys did not completely melt during the diffusion experiment because some segregated parts in the initial condition of Sn-3at.%Cu might have higher concentration with a higher liquidus temperature than 573 K. This study was aimed to investigate whether the samples of SnCu alloys did melt by measuring D_{CuSn} at the initial concentration of Sn-1at.%Cu, which is sufficiently lower than the concentration where the melting point of the SnCu alloy is 573 K. Furthermore, the concentration dependence of D_{CuSn} in the concentration region including the previous measurement condition[2)] was also investigated.

2. Experimental Method

The shear cell technique and the stable density layering[3)] were used. The diffusion couples of pure Sn and SnCu alloys were set in four capillaries. The initial concentration of Cu in the SnCu alloys was 1 at. %. The samples of SnCu alloys were placed on the lower side so as to provide a stable density layering for suppressing natural convection. After evacuating the chamber containing the shear cell device, the furnace was heated up to the diffusion temperature at 573 K. After homogenization, the diffusion process started. At the end of the diffusion time of 28800 s, the samples were divided into small cells mechanically and cooled down. The concentration of Cu of each cell was obtained.

3. Results

Figure 1 (a) shows the concentration profiles of the experiment using Sn-1at.%Cu. The impurity diffusion coefficients of Cu in liquid Sn D_{CuSn} were obtained from fitting concentration profiles to the thick layer solution. As the coefficients of determination R^2 were higher than 0.999 in all experiments, these fitting were done well. The average of four measured values of D_{CuSn} was 2.89×10^{-9} m^{2}s^{-1} and the relative standard derivation was 0.7 %. The reproducibility of the concentration profile was high because the variation in initial concentration was small and four concentration profiles almost agreed each other. In order to compare the concentration profiles of different initial concentrations, **Fig. 1 (b)** shows the normalized Cu concentration profiles of experiments using Sn-1at.%Cu and Sn-3at.% at 573 K. These were selected as an example from four normalized Cu concentration profiles for each initial concentration. From **Fig. 1 (b)**, the reproducibility of the concentration profile of the experiment using Sn-3at.%Cu was high because the normalized concentration profile of the experiment using Sn-3at.%Cu almost agreed with that of the experiment using Sn-1at.%Cu. Similarly, the others normalized concentration profiles of the experiment using Sn-3at.%Cu almost agreed with those of the experiment using Sn-1at.%Cu.



(a) (b)

Fig. 1 Cu concentration profiles at 573 K: (a) Sn-1at.%Cu and (b) normalized Cu concentration profiles of the experiments using Sn-1at.%Cu and Sn-3at.%Cu. The symbol "←g" indicates the direction of gravity.

4. Disccusion

The average of eight values of D_{CuSn} which were measured in this experiment and previous one[2] was 2.89×10^{-9} m^{2}s^{-1} and the relative standard derivation was 1.36 %. Therefore, D_{CuSn} has almost no concentration dependency and the samples of the SnCu alloys were confirmed to have melted during experiment using Sn-3at.%Cu. The obtained D_{CuSn} of 2.88×10^{-9} m^{2}s^{-1} was 16% smaller than the estimated value.

5. Conclusion

The samples of SnCu alloys were confirmed to have melted during the diffusion experiment using Sn-3at.%Cu because of the high reproducibility of the concentration profiles. D_{CuSn} has almost no concentration dependence and the obtained D_{CuSn}(573 K) of 2.88×10^{-9} m^{2}s^{-1} was confirmed to be reliable, which was 16 % smaller than the estimated value.

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 and conducted as a part of the research project in 2019 for Research Assistant (Masato SHIINOKI) in Kagami Memorial Research Institute for Materials Science and Technology, Waseda Univ.

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