The liquid diffusion measurements using the shear cell technique on the ground

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Abstract

In order to measure the liquid diffusion coefficients on the ground and to grasp the limitation of the ground experiments, the experimental method of diffusion was determined. It was based on the shear cell technique coupled with the techniques to make a thin and homogeneous sample and to analyze the diffusion sample.

Introduction

The liquid diffusion coefficients are important properties both for industries and for liquid theories. Although it is hard to measure the liquid diffusion coefficients on the ground because of the buoyancy convection, it can be measured with thin samples within a certain error if the temperature is not so high compared with the melting temperature. The highly precise liquid diffusion coefficients at high temperature is needed to clarify the diffusion mechanisms. Nevertheless, diffusion coefficients are useful in industrial fields, even though they are limited to be near the melting points and contain some error. We are developing the shear cell technique for the ground based experiments, and examining the limitation and the errors of the ground diffusion experiments. We established the techniques for ground diffusion experiments based on the shear cell, that is how to make thin samples and how to analyze the concentration, this year.

Outline of the diffusion experiments

Ag-Au system was selected for the diffusion experiments on the ground. Ag-Au is a complete solid solution system, in which the temperature difference between the solidus and the liquidus lines is narrow as shown in Figure 1. Especially the temperature difference at Au concentration of less than 5 at% is almost zero. The diffusion couple of Ag and $Ag_{0.95}Au_{0.05}$ was selected for the ground diffusion experiments. The merit of the selected sample is as follows:

1. The dendritic growth is hardly occurred because of the narrow temperature difference of the liquidus and the solidus lines. Therefore, homogeneous samples can be easily obtained and the diffusion sample after the shear cell experiments is expected to be homogeneous.

2. EPMA (Electron Probe Microanalyzer) in NASDA can be employed for the analysis. The accuracy is high enough for Ag and Au.

The experimental procedure is
(1) sample preparation,
(2) setting the samples in a shear cell,
(3) shear cell experiment and (4) sample analysis. The sample diameter of the shear cell was 1.0 mm. The shear cell method is described in Sec.2.(11).

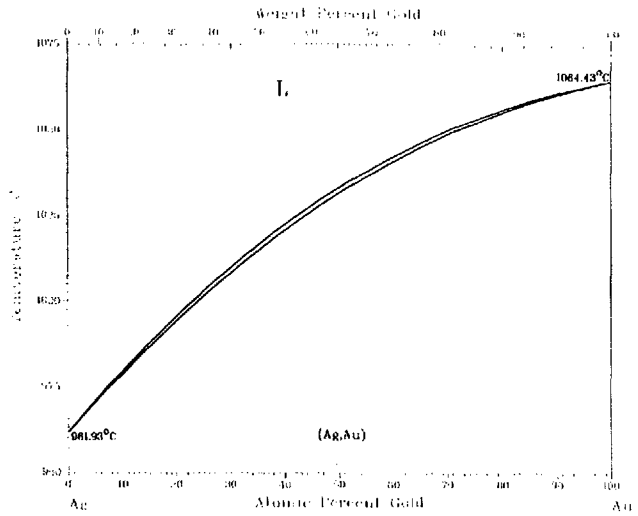


Figure 1 Phase diagram of the Ag-Au system (Binary Alloy Phase Diagrams Second Ed. (1996) ASM International)

How to make thin and homogeneous samples

First, the sample material is cleaned and the oxide of the sample material is removed by passing through a small hole (0.5 mm diameter). 7 mm diameter samples of Ag and $Ag_{0.95}Au_{0.05}$ are cast into a 1 mm diameter cylinder using a crucible as shown in Figure 2. The crucible can be divided into two parts along the axial plain and the sample can be easily taken out from the crucible. The concentration distribution in the cylindrical sample of $Ag_{0.95}Au_{0.05}$ with 1 mm diameter was analyzed and the homogeneity was confirmed within 0.2 at%.

Diffusion experiment with the shear cell

Diffusion experiments were performed at 1273 K for 4 minutes using the Large Isothermal Furnace. The diffusion couple of Ag and $Ag_{0.95}Au_{0.05}$ was joined and kept at 1273 K for 4 minutes, and was divided into 8 pieces with the thickness of 1.5 mm. The sample was kept further more at the diffusion temperature for 6 minutes after the sample deviation in order to homogenize the divided samples in each cell. The concentration distribution of the samples in each cell was analyzed at both sides and the homogeneity in each sample was confirmed within 0.1 at%.

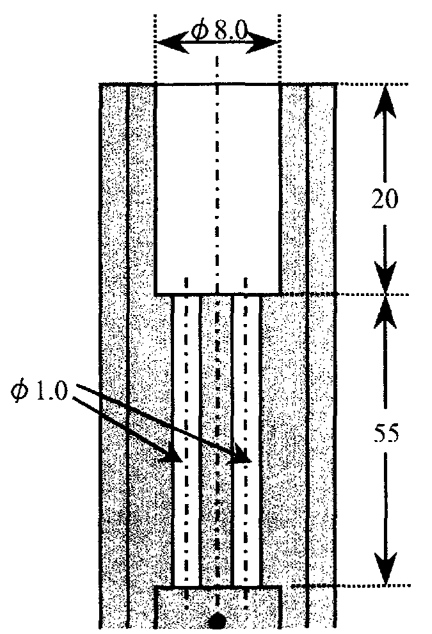


Figure 2 Crucible for casting a cylindrical sample with 1.0 mm diameter

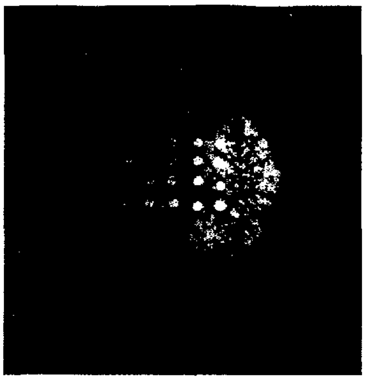
How to analyze the diffusion sample

In order to analyze the diffusion samples in each cell, they were fixed into holes in a graphite holder with a carbon paste and were polished up to #1200 as shown in Photograph 1. The samples were analyzed by EPMA (Electron Probe Microanalyzer) at 20 points each. The scatter in the 20 points was less than +/-0.1 at%. The diffusion coefficient was calculated from the concentration distribution obtained from the EPMA analysis by fitting the analytical solution of the Fick's second law. The Fick's second law is

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x}\left(D\frac{\partial C}{\partial x}\right). \qquad (1)$$

Here, C is the concentration and D is the diffusion coefficient. The analytical solution for the diffusion couple, which is long enough to keep the initial concentration at the ends of the couples, is

$$C(c,t) = \frac{C_2 - C_1}{2}\left\{1 - erf\left(\frac{x}{2\sqrt{Dt}}\right)\right\} + C1. \qquad (2)$$



Photograph 1
The diffusion samples fixed in a graphite holder for EPMA analysis

Here, C_1 and C_2 are the initial concentrations of the each sample piece. The error function, *erf*, is defined as

$$erf(z) = \frac{2}{\pi^{1/2}}\int_0^z \exp\left(-\eta^2\right)d\eta. \qquad (3)$$

Figure 3 shows the concentration distribution and the fitting curve to Fick's second law. The diffusion coefficient was obtained as 3.0×10^{-9} m^2/s at 1273 K.

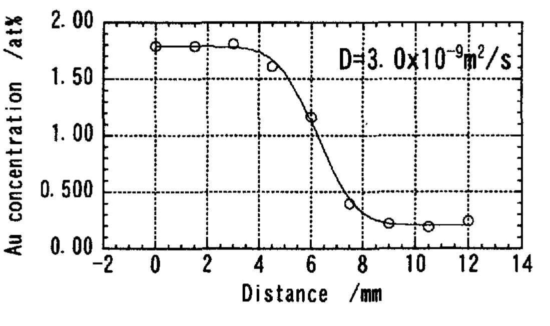


Figure 3 Concentration distribution after the diffusion experiment at 1273 K

Conclusions

The method of diffusion measurements in melts on the ground was discussed. The method is based on the shear cell method coupled with the method of preparation of a thin sample, concentration analysis and evaluation of diffusion coefficient. We will clear the limitation of accuracy of ground experiments in the near future.