Development of shear cell with X-ray observation system

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ABSTRACT

The most effective way for liquid diffusion measurements is the shear cell technique. In this method, a diffusion couple is only joined at a target temperature to avoid diffusion during heating and cooling processes, and to avoid solidification effects. Numerical simulation confirmed that the misalignment of a diffusion couple hinders precise diffusion experiments. Hence, the liquid sample should be observed during the shear cell experiments to confirm the correct alignment. A direct X-ray observation system was thus specifically developed and the impurity diffusion of Au in liquid Ag was successfully measured using this apparatus.

1. INTRODUCTION

The liquid diffusion coefficient is an important property for scientific endeavor, in particular as the many-body problems in the fundamental science, and for industrial applications (e.g. crystal growth of semiconductors). However, it is difficult to measure it on the ground because the buoyancy convection is hardly avoided. One elegant way to circumvent this problem is to carry out experiments in microgravity. Under the microgravity environment offered by the Space Shuttle and the International Space Station, buoyancy convection can be suppressed and the atomic motion in liquids is governed only by diffusion. It was reported that diffusion coefficients obtained by microgravity experiments were smaller than those obtained on the ground. Moreover, accurate coefficients were expected to be measured under reduced gravity conditions [1,2].

Diffusion coefficients are often measured by the long capillary method, in which two pieces of samples with different concentrations are put into contact, heated, kept at a specific temperature, and solidified [3]. The diffusion coefficient is determined from the concentration distribution in the solidified sample. This method faces two problems for liquids. First, the concentration distribution includes not only the diffusion at the target temperature but also the contribution of heating and cooling processes. Second, the solidification spoils the concentration distribution due to shrinkage and segregation [4]. The shear cell technique provides a solution to eliminate these problems [5]. In this technique, each piece of a diffusion couple is separated during heating process and joined after the temperature is stabilized (Fig. 1). The diffusion sample is then divided into small pieces before cooling and solidifying. As evidenced by our numerical simulation, it is important to keep the diffusion couple correctly aligned in order to conduct precise diffusion measurements [6]. The misalignment should be less than 10 % of the sample diameter to obtain diffusion coefficients from space experiments with accuracy better than 3 %. A misalignment of 10 % may easily occur due to the small diameter of the liquid column itself. However, a shear cell experiment is usually carried out without confirming how the diffusion couple is joined. Therefore, we developed a shear cell technique with an X-ray observation system. By using this apparatus, diffusion experiments were performed with observation of samples.

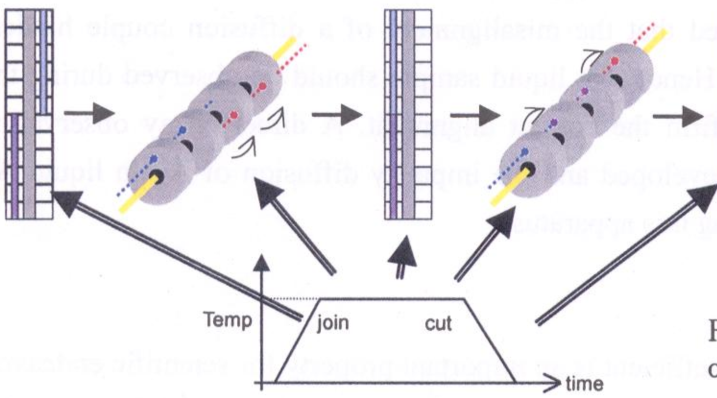


Figure 1 Schematic drawing of shear cell technique

2. EXPERIMENTAL

The shear cell consisted of a pile of disks, a rotation shaft and a key bar to determine the position of each disk (Fig. 2). Two diffusion samples of 1.0 mm diameter and 30 mm length overall (15 mm for each piece of a diffusion couple) were set through 21 disks. The thickness of each disk was selected as 1.0 mm since the disturbance length by the flow on shearing was found to be almost the same size as the sample diameter [7]. The disks were made of glassy carbon (Nisshinbo Industries, Inc.), which is a dustless and hard material. A sample cartridge consisted of the shear cell

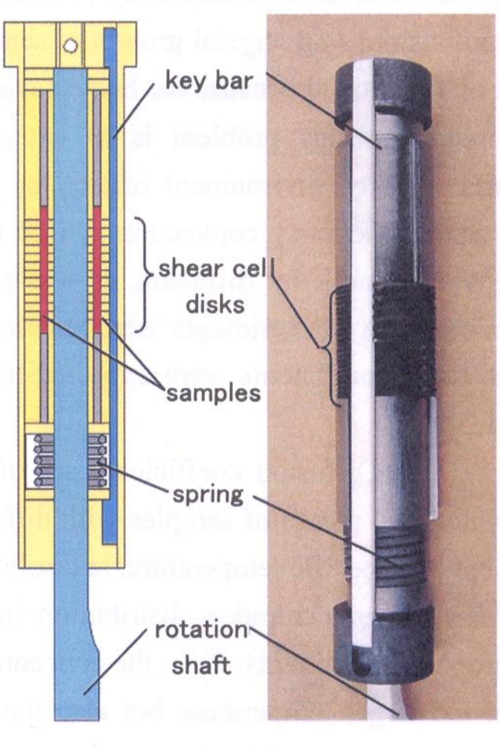


Figure 2 Shear cell configuration

described above, a ceramic heater, a sapphire or a quartz cartridge to keep vacuum, and a motor to rotate the cell (Fig. 3). Since the part of the cartridge around the samples needed to be X-ray transparent, the heater was made of pyrolytic boron nitride and pyrolytic carbon (Advanced Ceramics International Corporation). The cartridge was set in an X-ray observation system, the AFEX (Advanced Furnace for microgravity Experiments with X-ray radiography) designed for the International Space Station experiments [8]. It comprised two sets of X-ray generator – detector and the samples could be observed from two directions, 0° and 60°.

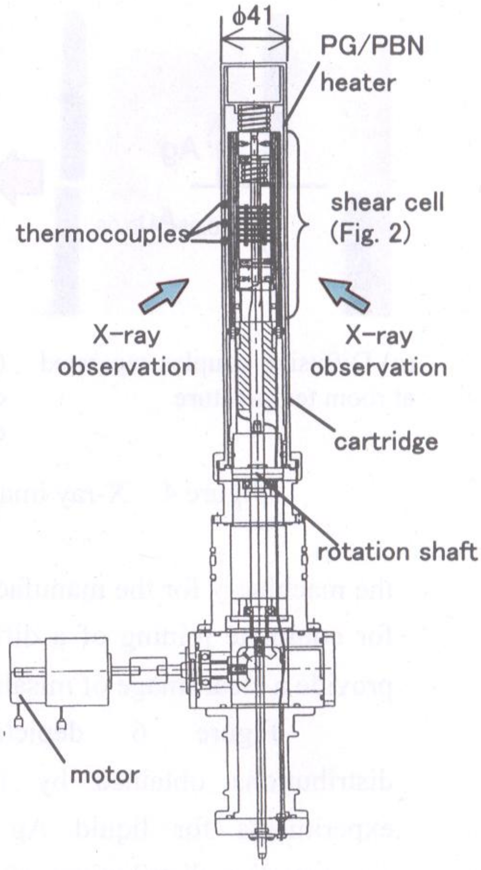
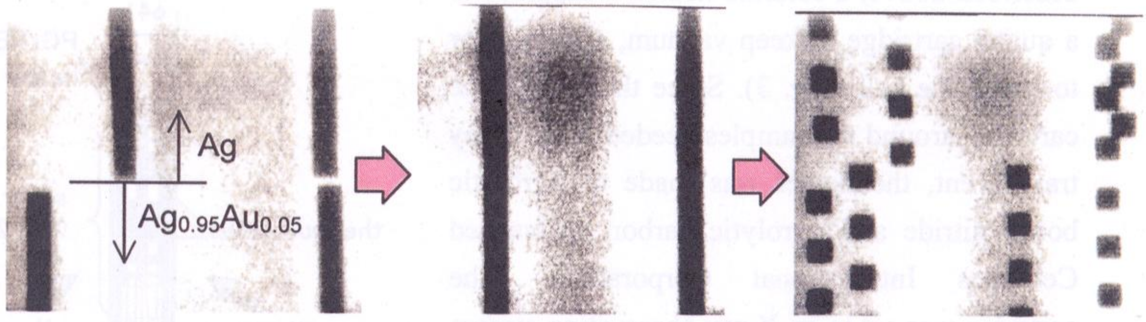


Figure 3 Shear cell cartridge for X-ray observation

An impurity diffusion coefficient of Au in Ag was measured using this shear cell. The sample was a diffusion couple of Ag and $Ag_{0.95}Au_{0.05}$, whose diameter was chosen to be 1.0 mm to suppress the buoyancy convection. The sample temperature was measured by three thermocouples, which were located on a crucible at the top, the middle and the bottom position of the sample (Fig. 3). The temperature was kept higher at the top to avoid the buoyancy convection. The concentration was analyzed by EPMA (Electron Probe Microanalyzer), whose accuracy was about 0.1 at% for Au and Ag.

3. RESULTS AND DISCUSSION

Two diffusion experiments were carried out at 1273 K for 4 and 10 minutes. A diffusion couple was created by the shaft rotation of 45° at first, keeping it for a while to stabilize the sheared flow, and finally further rotation of 20°. This two stage rotation was adopted as it reduced induced flow in the liquid sample by minimizing rotation. After the diffusion process finished, the liquid sample was divided into small pieces by a 120° rotation. The samples were clearly observed by X-ray during the experiments, as shown in Fig. 4. This system confirmed that the liquid samples were joined without misalignment and were divided into small pieces successfully (Fig. 4(b)). Figure 5 shows joined samples with a misalignment of about 200 μm (20 % of sample diameter). Such a misalignment easily occurs even in machinery within 50 μm accuracy if the shear cell disks are manufactured without special care. It was found that the accuracy of



(a) Diffusion couples separated at room temperature (b) Diffusion couples joined completely and diffusion experiments carried out (c) Diffusion samples divided into small pieces and cooled down

Figure 4 X-ray image of samples during diffusion experiments

the machinery for the manufacturing of the shear cell should be less than a few tens μm for complete joining of a diffusion couple. The present X-ray imaging system could provide a clear image of misalignment within 50 μm.

Figure 6 depicts the concentration distributions obtained by the present shear cell experiments for liquid Ag – $Ag_{0.95}Au_{0.05}$. These concentration distributions after the experiments were in accordance with Fick's second law. The diffusion coefficients were calculated for complete joined samples as 13.3 x 10^{-9} m^2 s^{-1} and 7.0 x 10^{-9} m^2 s^{-1} respectively for 4 and 10 minute experiments. In the shorter experiment, the flow on shearing influences significantly and the obtained diffusion coefficient

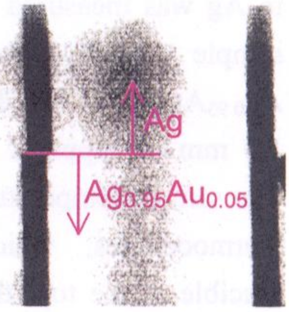
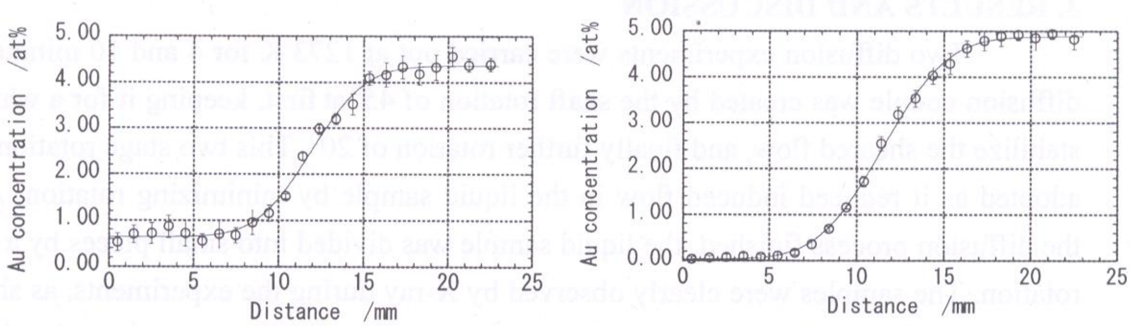


Figure 5 Joined sample with misalignment due to machinery inaccuracy



(a) Diffusion time = 240 s : diffusion coefficient = 13.3 x 10^{-9} m^2 s^{-1}

(b) Diffusion time = 600 s diffusion coefficient = 7.0 x 10^{-9} m^2 s^{-1}

Figure 6 Concentration distributions after experiments at 1273 K became larger. It is important to grasp the optimum relation between the rotation rate and the diffusion time to conduct accurate shear cell diffusion measurements under microgravity.

5. CONCLUSIONS

A shear cell coupled with an X-ray imaging system was developed and diffusion experiments were carried out. From the direct X-ray observation, it was confirmed that the diffusion couples could join completely, without any misalignment, and could be divided into many pieces successfully.

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