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ジルコニウム融体の表面張力と酸素溶解量の時間依存性に対する雰囲気ガス種の影響

Influence of Dissolved Oxygen from Different Atmospheric Gas on Surface Tension of Liquid Zirconium

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1. Introduction

Surface tension and its temperature coefficient play a key role in high temperature processes with a significant ratio of free surface to volume such like a welding. For example, surface tension is required to discuss a wetting behavior of the melt. Temperature coefficient of surface tension is a driving force of Marangoni convection affecting mass/heat transportation in melt. However, the reported data of surface tension of high temperature liquid metals often shows a scattering. One of the possible reasons for the scattering is that it is difficult to suppress a contamination of the sample from a supporting material of the sample as long as long as the measurement was carried out by using conventional container methods. Furthermore, influence of oxygen partial pressure of measurement atmosphere (P_{O2}) on the surface tension has not been considered in many studies though oxygen acts as one of the strongest surfactants for metallic melt. Especially when the sample has a high oxygen solubility like zirconium, the amount of dissolved oxygen can increase with time during the measurement.

In this study, liquid zirconium was maintained at high temperature under levitated state by using electromagnetic levitator (EML). We investigated a time dependence of the amount of dissolved oxygen in liquid zirconium and the corresponding surface tension under different gas atmosphere. The purpose of this investigation was to measure an accurate surface tension of liquid zirconium free of contamination from container even at high temperature sufficiently above the melting point in consideration of influence of P_{O2}.

2. Experimental Procedure

A cubic zirconium with a purity of 99.2 mass % was chemically cleaned by a fluonitric acid solution using an ultrasonic cleaning machine and then placed on a quartz holder in the chamber of EML. The sample was electromagnetically levitated and then melted under the flow condition of the atmospheric gases at 2 L/min. The P_{O2} of the atmospheric gas was fixed at 10^{-7} atm under high purity mixture gas of Ar-He, and at 10^{-9} and 10^{-11} atm under gas phase equilibrium between H_2 and CO_2, which was confirmed by zirconia type oxygen sensor operated at 1008 K installed at the inlet of chamber. A semiconductor laser beam was also used to heat the sample to a sufficiently high temperature. The temperature of levitated droplet was controlled by varying the output power of the laser and the partial pressure of argon and helium gases. The oscillation behavior of the droplet was observed from above using a high-speed video camera. The frequencies of the surface oscillations of the m= 0, ±1, and ±2 for the l=2 mode and the center of gravity were analyzed from time-sequential data of the HSV images through fast Fourier transformation (FFT). Surface tension of the liquid zirconium was calculated from these frequencies using the Rayleigh equation [1] and Cummings and Blackburn calibration [2]. The oxygen concentration of the sample after the surface tension measurement was analyzed by an inert gas fusion oxygen analyzer.

3. Results and Discussion

Figure 1 shows the time dependence of the amount of dissolved oxygen in liquid zirconium and corresponding the surface tension, when it was maintained at 2300 K under flow condition of high purity Ar-He and Ar-He-H_2-CO_2 gases. When the Po_2 is controlled at 10^{-7} atm using Ar-He gas (●), no variation is detected in the amount of dissolved oxygen and corresponding the surface tension even after 20 minutes.

However, the amount of dissolved oxygen increases with time under Ar-He-H_2-CO_2 gas atmosphere even though the Po_2 of the gas is decreases to ~10^{-9} atm (■) and ~10^{-11} atm (▲). This is because the chemical equilibrium between sufficient volumes of H_2 and CO_2 gases forms oxygen gas immediately depending on a consumption of oxygen gas of atmosphere through the dissolution in liquid zirconium. The oxygen potential in liquid zirconium is always lower than that in the H_2-CO_2. Oxygen gas supply rate through the buffer effect of H_2-CO_2 gas would be much higher than that by simple gas mixture of Ar-He gas of 2 L/min.

The surface tension of liquid zirconium decreases depending on the increase in the amount of dissolved oxygen under Ar-He-H_2-CO_2 gas atmosphere. These results indicate that surface tension of liquid zirconium shows a time dependence induced by a variation of the amount of dissolved oxygen when oxygen buffer gas such as H_2 and CO_2 is used as a measurement atmosphere.

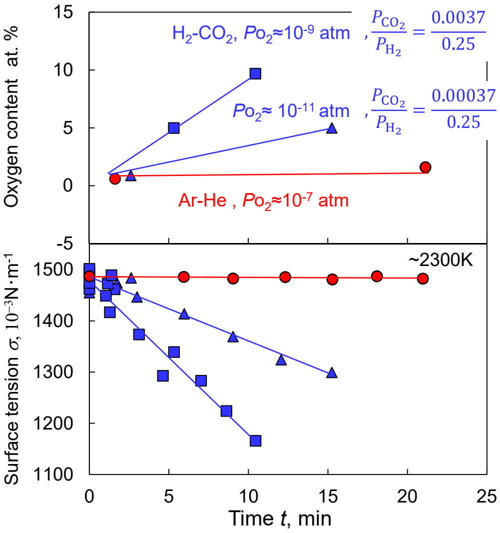

Fig. 1 Variation of oxygen content and surface tension of zirconium melt with time under atmospheric gases of Ar-He (Po_2≈10^{-7} atm :●) and H_2-CO_2 (Po_2≈10^{-11} atm :▲, ≈10^{-9} atm :■) .

Acknowledgement

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References

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2) D. L. Cummings, D. A. Blackburn: J. Fluid Mech., 224, (1991) , p. 395.