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Zr 融体の表面張力に及ぼす温度と酸素の影響

Influence of Temperature and Oxygen Concentration on Surface Tension of Molten Zirconium

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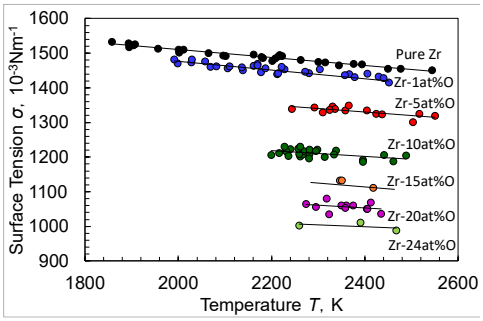
Abstract:
The surface tension of molten Zr–O alloys was measured as a function of temperature and composition using the oscillating droplet method with an electromagnetic levitation furnace, as a ground-based preliminary experiment for an upcoming microgravity study using the Electrostatic Levitation Furnace (ELF) onboard the International Space Station (ISS). The surface tension decreased with increasing temperature, as observed in pure molten zirconium. An increase in oxygen content decreased the surface tension of the molten samples. No detectable variation in oxygen content was observed during the measurements conducted in a mixed argon-helium atmosphere at an oxygen partial pressure of approximately 10^{-2} Pa.



Keywords: Surface tension, High temperature melt, Oxygen dissolution, Electromagnetic levitation

1. Introduction

The microgravity experiment mission, named OASIS, is planned to be conducted on the International Space Station (ISS) using the Electrostatic Levitation Furnace (ELF), with the aim of measuring accurate and precise thermophysical properties including density, surface tension, and viscosity. Zirconium alloys are widely used in products such as nuclear reactors and chemical processing plants due to their excellent corrosion resistance and low thermal neutron adsorption cross section. Because liquid zirconium exhibits a remarkably high oxygen solubility, the oxygen content can increase over time during measurements. Such an increase can influence the results—particularly surface tension—since oxygen is among the most potent surfactants.

In the present study, the surface tension of molten Zr–O alloys was measured as a function of temperature and composition using the oscillating droplet method with an electromagnetic levitator (EML), as a ground-based preliminary experiment for an upcoming OASIS mission.

2. Experimental Procedure

Zr-O alloys with oxygen contents of 1–20 at% were prepared by arc melting from pure zirconium (99.6 at% purity; 0.5at%O) and ZrO_2 (98.6 at% purity). The resulting alloys were cut into a cubic piece weighing approximately 1000 mg with a side length of 6 mm. The alloys were chemically cleaned in a fluoronitric acid solution using an ultrasonic cleaner, and subsequently rinsed with acetone. After the cleaned sample was then placed on a quartz holder positioned within the levitation coil in EML, the EML chamber was thoroughly

purged with high purity argon gas with an oxygen partial pressure (P_{O_2}) of approximately 10^{-2} Pa. The P_{O_2} of the gas was monitored at the inlet of the chamber using a zirconia oxygen sensor operated at 1008 K. The sample was electromagnetically levitated and then melted under an argon gas flow of 1 L・min. A semiconductor laser beam was used to heat the levitated droplet sufficiently above its melting point.The temperature of the levitated droplet was controlled by adjusting the laser output power, and monitored using a monochromatic pyrometer.

After the sample temperature and P_{O_2} stabilized, the oscillation behavior of the droplet was observed from above using a high-speed video (HSV) camera. The frequencies of the surface oscillations of the m = 0, ±1, and ±2 for the l = 2 mode and those of the center of gravity were analyzed from the time-sequenced data of the HSV images using the fast Fourier transformation (FFT). The surface tension of molten Zr–O sample was calculated from these frequencies using the Rayleigh equation[1], calibrated with the Cummings and Blackburn calibration [2]. The oxygen contents of the sample after the surface-tension measurement was analyzed using an inert gas fusion oxygen analyzer (EMGA-930, HORIBA Ltd.).

3. Results and Discussion

Figure 1 shows the surface tension of molten Zr–O samples measured by EML at an argon gas flow rate of 1 L・min^{-1} as a function of temperature and oxygen content. Under this gas flow condition, no significant change in oxygen content was detected after the surface tension measurement, even though the P_{O_2} of 10^{-2} Pa in the gas is higher than the equilibrium value for the formation of ZrO_2(s). This is attributed to the gas flow rate being the rate-determining step in the dissolution of atmospheric oxygen into molten samples, as in the cases of molten zirconium and titanium.

The surface tension of the molten samples decreases linearly with increasing temperature, independent of oxygen concentration. Increasing oxygen content lowers the overall surface tension and reduces the magnitude of the temperature coefficient. This reduction in the temperature coefficient is attributed to the surface oxygen coverage approaching saturation, which stabilizes the surface structure and diminishes the temperature-induced increase in surface entropy. According to the Butler model, preferential adsorption of oxygen alters the surface composition, and when adsorption follows a Langmuir-type isotherm, surface coverage increases rapidly at low bulk concentrations but levels off near saturation. In this regime, further increases in oxygen content have little effect on the temperature dependence of surface tension because the configurational contribution to the surface entropy is already minimized.

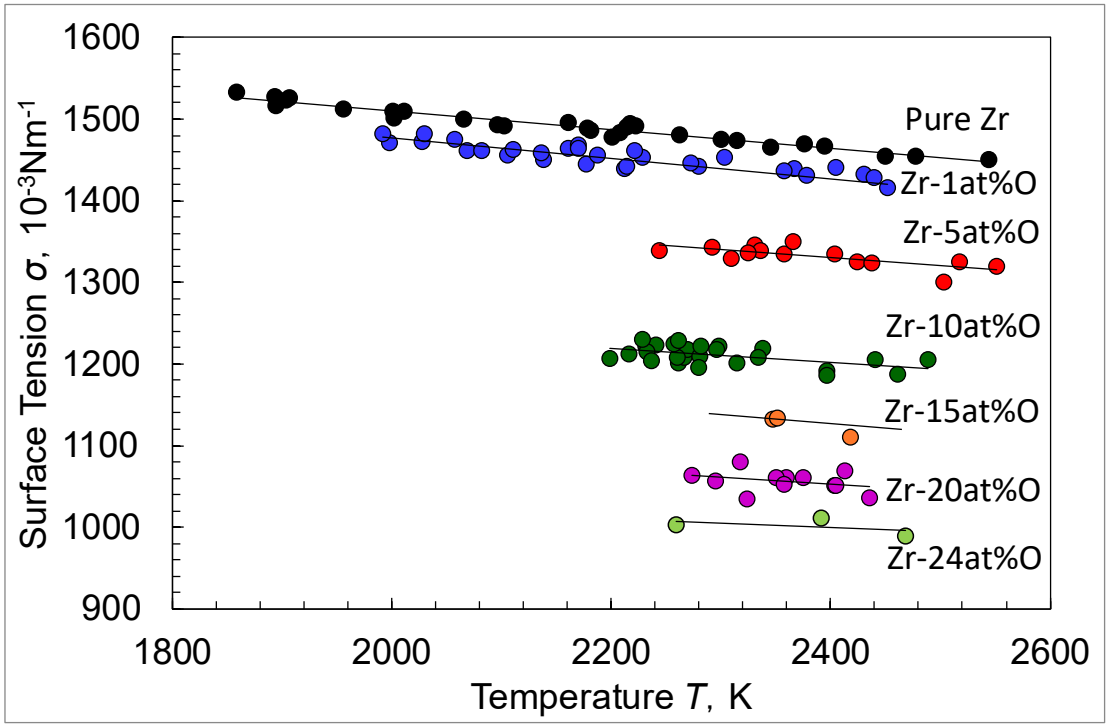


Figure 1. Surface tension of molten Zr-O samples measured by EML

4. Conclusion

Surface tension of molten Zr–O alloys decreases linearly with temperature, with higher oxygen contents lowering both absolute values and temperature coefficients. These results support the design and interpretation of the upcoming OASIS microgravity experiments.

Acknowledgments

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