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電磁浮遊法を用いた二元系合金融体の表面張力測定

Surface Tension Measurements of Molten Iron–Based Binary Alloys Using Electromagnetic Levitator

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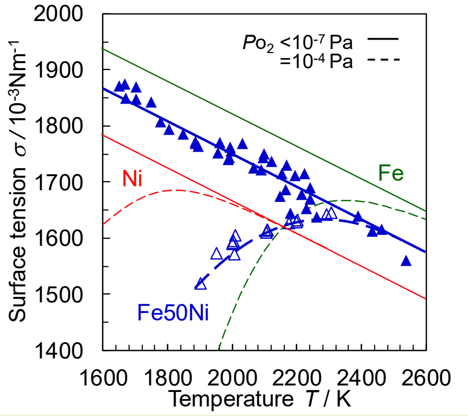
Abstract: We measured the surface tension of molten Fe–Ni alloys under containerless conditions using the oscillating droplet method in an electromagnetic levitation furnace. This study investigated the effects of temperature, alloy composition, and oxygen adsorption on the surface tension. Under low oxygen partial pressure (P_{O_2}), the surface tension of molten Fe–Ni alloy closely matched the weighted average of molten pure Fe and Ni regardless of alloy composition. At high P_{O_2}, oxygen adsorption led to a decrease in surface tension at lower temperatures, whereas oxygen desorption caused it to increase with temperature. Under the same P_{O_2} conditions, molten Fe-rich alloys exhibited surface tension values close to those of molten pure Fe, whereas molten Ni-rich alloys showed values close to those of molten pure Ni, reflecting the distinct oxygen affinities of Fe and Ni. The molten Fe–50 at% Ni alloy consistently exhibited surface tension values near the weighted average of those of molten pure Fe and Ni under all P_{O_2} conditions. These results suggest that in molten Fe–Ni alloys, the element with the higher concentration dominates the surface composition, and its interaction with adsorbed oxygen significantly influences the surface tension.



Keywords: Oxygen adsorption, High-temperature melt, Surface tension, Fe-Ni alloy, Electromagnetic levitation

1. Introduction

Surface tension is one of the most important thermophysical properties for improving and optimizing high-temperature melt processes such as welding and casting, as it affects the melt surface shape and drives Marangoni convection. To accurately measure the surface tension of a high-temperature melt, it is crucial to prevent contamination of the sample from supporting materials, as even trace amounts of impurities can significantly alter the surface tension[1)]. Atmospheric oxygen adsorption also influences surface tension, since oxygen acts as one of the strongest surfactants in metallic melts[2)]. Recently, accurate surface tension measurements of several pure molten metals have been conducted using electromagnetic levitation (EML), which enables contamination-free conditions and allows the effects of oxygen adsorption to be assessed. However, the effects of oxygen adsorption on the surface tension of molten alloys remain insufficiently understood.

In this study, we measured the surface tension of molten Fe–Ni binary alloys, which have an activity coefficient $\gamma \approx 1$, under containerless conditions using the oscillating droplet method with an EML. This study aimed to provide fundamental data on how temperature and oxygen adsorption affect the surface tension of molten alloys.

2. Experimental procedure

An ingot of Fe–Ni alloy was prepared from high-purity iron and nickel, each with a purity of 99.99 mass% or higher. The ingot was cut into a nearly cubic shape with a mass of approximately 1 g, and then chemically etched in a Nital solution using an ultrasonic cleaner, followed by rinsing with acetone. The sample was electromagnetically levitated and melted in a flowing mixed-gas atmosphere of Ar–He–H_2–CO_2 at a rate of 2 L·min^{-1}. The oxygen partial pressure of the gas (P_{O_2}) was controlled through the gas-phase equilibrium between H_2 and CO_2. To confirm the P_{O_2} of the inlet gas, a zirconia-type oxygen sensor was used, operating at 1008 K, considering the temperature dependence of P_{O_2}. The droplet temperature was controlled by adjusting the partial pressures of argon and helium gases, which have different thermal conductivities, using a monochromatic pyrometer. After the indicated temperature and P_{O_2} values stabilized, the oscillation behavior of the droplet was observed from above using a high-speed video (HSV) camera.

The frequencies of surface oscillations of $m = 0$, ±1, and ±2 for the $l = 2$ mode, and the center of gravity were analyzed from sequential HSV images using fast Fourier transformation (FFT). The analysis also accounted for apparent droplet rotations induced by phase differences between $m = +1$ and $m = -1$, and $m = +2$ and $m = -2$ components, in addition to real rotational effects [3,4)]. The surface tension of the molten Fe–Ni alloy was calculated from these frequencies using the Rayleigh equation[5)] and the calibration by Cummings and Blackburn[6)].

3. Results and Discussion

Figure 1 shows the temperature dependence of the surface tension of molten Fe–25 at% Ni, Fe–50 at% Ni, and Fe–75 at% Ni alloys, measured by EML under two different gas atmospheres: Ar–He–H_2 (P_{O_2} < 10^{-7} Pa) and Ar–He–H_2–CO_2 (P_{O_2} = 10^{-4} Pa). Data for molten pure Fe and Ni are also included for comparison. When P_{O_2} is reduced to below 10^{-7} Pa under the Ar–He–H_2 gas mixture, the surface tension of molten Fe–Ni alloy decreases linearly with increasing temperature regardless of the alloy composition, similar to the behavior observed in molten pure Fe and Ni. The measured values closely follow the composition-weighted average of the surface tension of molten pure Fe and Ni.

In contrast, P_{O_2} is increased to 10^{-4} Pa under the Ar–He–H_2–CO_2 gas mixture, the surface tension of molten Fe–25 at% Ni alloys exhibits a convex-upward temperature dependence. This behavior is attributed to oxygen adsorption at low temperatures, which lowers the surface tension followed by oxygen desorption at higher temperatures, resulting in a temporary increase in surface tension. As temperature further increases

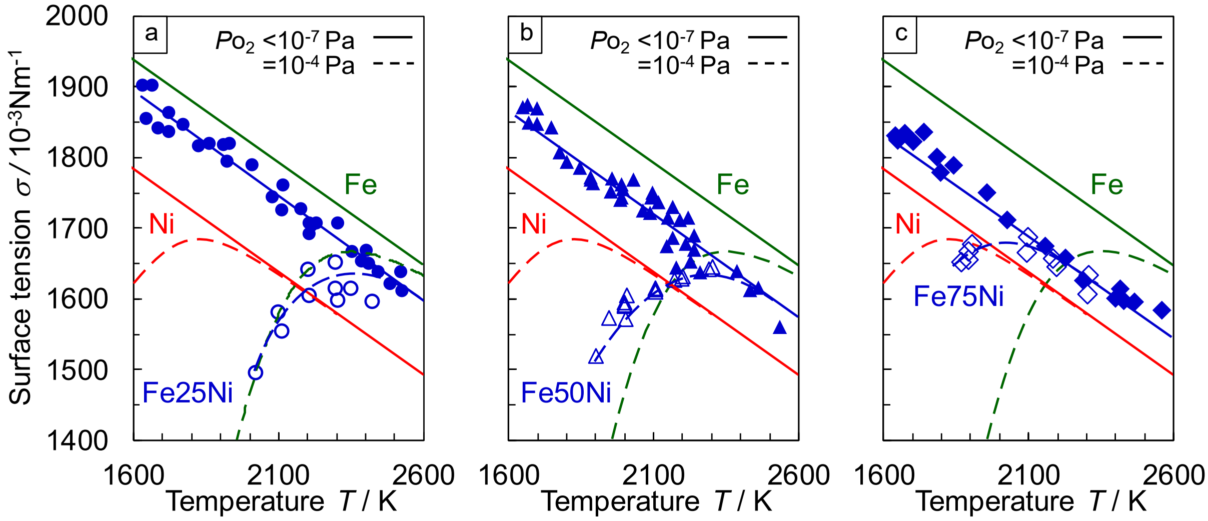


Figure 1. Temperature dependence of surface tension of molten Fe–Ni alloys with various Ni compositions, measured at P_{O_2} = 10^{-4} Pa: (a) Fe–25 at% Ni, (b) Fe–50 at% Ni, (c) Fe–75 at% Ni.

and oxygen adsorption becomes less effective, the surface tension begins to decrease again, similar to the behavior observed under low P_{O_2} conditions.

Even under oxygen adsorption, the surface tension of molten Fe–50 at% Ni alloy consistently remains close to the composition-weighted average of molten pure Fe and Ni. However, when oxygen is adsorbed at low temperatures, the surface tension of molten Fe–25 at% Ni alloy tends to be closer to that of molten pure Fe, while that of molten Fe–75 at% Ni alloy approaches the values for molten pure Ni, rather than the composition-weighted average. These results suggest that oxygen adsorption alters the surface composition of molten Fe–Ni alloys depending on the relative oxygen affinities of Fe and Ni. As a result, Fe tends to dominate the interfacial behavior in Fe-rich compositions, whereas Ni becomes dominant in Ni-rich alloys.

4. Summary

The surface tension of molten Fe–Ni alloys was measured using the oscillating droplet method with EML, and the effects of temperature, alloy composition, and P_{O_2} were investigated. Under low P_{O_2} conditions, where oxygen adsorption is negligible, the surface tension of molten Fe–Ni alloy closely followed the composition-weighted average of those of molten pure Fe and Ni. In contrast, under high P_{O_2} conditions where oxygen adsorption occurs, the surface tension was influenced by the surface composition: it was dominated by Fe in Fe–rich alloys and by Ni in Ni–rich alloys.

Acknowledgments

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Conflicts of Interest

The authors declare no conflict of interest.

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