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Surface tension of molten iron measured by oscillating droplet method using electromagnetic levitation[†]

—Influence of oxygen adsorption on surface tension—

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1. Introduction

Surface tension is one of the most important thermophysical properties to understand and optimize in a welding process through numerical simulation because its temperature coefficient is a driving force of Marangoni convection which affects the shape of a weld pool dramatically [1]. An accurate surface tension and its temperature coefficient at high temperature melt are strongly required.

The surface tension of molten metals is considerably influenced by oxygen partial pressure, $P\mathrm{o}_2$, of the ambient atmosphere because oxygen which acts as surfactant is adsorbed to a melt surface from the atmosphere gas [2]. Thus it is very crucial to consider the effect of $P\mathrm{o}_2$ on surface tension measurement. When $P\mathrm{o}_2$ is low, the surface tension of molten metal generally shows a negative temperature coefficient. In this case the Marangoni convection flows radially outward in a weld pool (**Fig. 1**) [1]. However, the temperature coefficient of surface tension increases with increasing $P\mathrm{o}_2$. Consequently it turns to a positive value as long as the molten metal is not oxidized under comparatively high $P\mathrm{o}_2$. In this case, Marangoni convection flows radially inward in a weld pool. Even when $P\mathrm{o}_2$ is high, the temperature coefficient is changed from a positive value to a negative value like a "boomerang shape" at higher temperature because the temperature elevation induce desorption of oxygen through an adsorption equilibrium mechanism. Although the boomerang shape temperature dependence of surface tension is expected for molten iron, it has not been experimentally confirmed yet because a conventional technique for a surface tension measurement such as the sessile drop method can assure only the measurement at a comparatively low temperature to prevent the contamination of the sample by the measurement device.

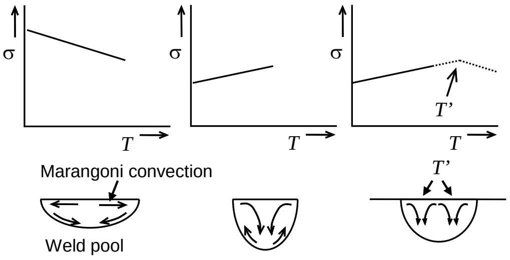

Fig. 1 relationship between temperature coefficient of surface tension and shape of weld pool

In this study the surface tension of molten iron was measured by an oscillating droplet method using electromagnetic levitation (EML). This technique enables us to measure the surface tension of molten metals over a wide temperature range including undercooling condition [3]. This technique can assure the $P\mathrm{o}_2$ control of the measurement atmosphere. The purpose of this study was to measure an accurate surface tension and its temperature coefficient for high temperature molten iron in consideration of oxygen partial pressure dependence at a wide temperature range.

2. Experimental procedure

About 800mg of an electrolytic iron (purity: 99.99%) was electromagnetically levitated and then melted under the flow condition (2L/min) of the mixed gas of high purity argon and helium to fix the $P\mathrm{o}_2$ of 10^{-2}Pa. The sample was also melted under flow condition of the Ar/He-5%H_2 mixed gas with a moisture content of 2.66 ppm to lower the $P\mathrm{o}_2$ by condensing of the H_2O formed from the following reaction;

$$H_2+\frac{1}{2}O_2 \rightarrow H_2O \qquad (1)$$

The $P\mathrm{o}_2$ of the inlet gas was confirmed by a zirconia oxygen sensor maintained at 1008K. The oscillation behavior and the temperature of the droplet were observed from the upper side using a high speed video camera and a pyrometer. The temperature of the sample was controlled by changing the flow rate of Ar and He gases.

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The frequencies of the surface oscillations of the $m = 0$, ±1, and ±2 for the $l = 2$ mode were analyzed from the observed images in consideration of the influence of thetwo types of droplet rotations, i. e. real rotation and apparent rotation [4]. The surface tension of molten iron was calculated from the Rayleigh equation [5] calibrated by the Cummings and Blackburn equation [6].

3. Results and discussions

When Ar/He-5%H_2 gas was used for the measurement, the oxygen sensor detected the Po_2 of 2.0×10^{-23}Pa at the inlet. However, Po_2 varies depending on the sample temperature because the equilibrium constant of the reaction (1) shows temperature dependence; the zirconia oxygen sensor can not measure the Po_2 of the gas surrounding the droplet maintained at certain temperature but it detects the Po_2 of an atmosphere introduced into the oxygen sensor operated at 1008K. The Po_2 of the Ar/He-5%H_2 gas was evaluated as a function of temperature using the standard Gibbs energy of formation of H_2O, in which the equilibrium constant of the reaction (1) is calculated from the Po_2 of 2.0×10^{-23}Pa of the inlet gas at 1008K measured by the oxygen sensor.

Figure 2 shows surface tension of molten iron measured by the oscillating droplet method using EML under the flow condition of mixed gases of Ar/He (Po_2) and Ar/He-5%H_2 [7]. Uncertainty for the measurement plot was calculated based on the GUM (ISO Guide to the Expression of Uncertainty in Measurement) [8], in which the coverage factor of k_p=2 was selected.

We successfully measured the surface tension of molten iron over a very wide temperature range over 780K including undercooling regions regardless of measurement atmosphere. When the Po_2 of the measurement atmosphere is fixed at 10^{-2} Pa regardless of sample temperature under the flow condition of high purity Ar and He gases, we succeeded in the experimental observation of a “boomerang shape” temperature dependence of surface tension of molten iron involving Po_2 for the first time; the surface tension increases as the sample temperature rises up to about 2150K, and then it decreases above this temperature because the Po_2 becomes less effective due to the temperature dependence of oxygen adsorption equilibrium constant. The pure surface tension of molten iron free from oxygen adsorption can be deduced from the negative temperature coefficient observed above 2150K described as a dotted line.

When the measurement of the surface tension is carried out under the flow condition of mixed gas of Ar/He-5%H_2, the surface tension decreases as the sample temperature rises. However, it does not appear to change with temperature uniformly but to show a kink at around 1850K. Since the variation of the surface tension at this kink is beyond the uncertainty of the measurement, it is not the scattering of the measurement originating from the measurement accuracy. This anomalous temperature dependence of the surface tension can be explained by the temperature dependence of the Po_2 of the mixed gas of Ar/He-5%H_2.

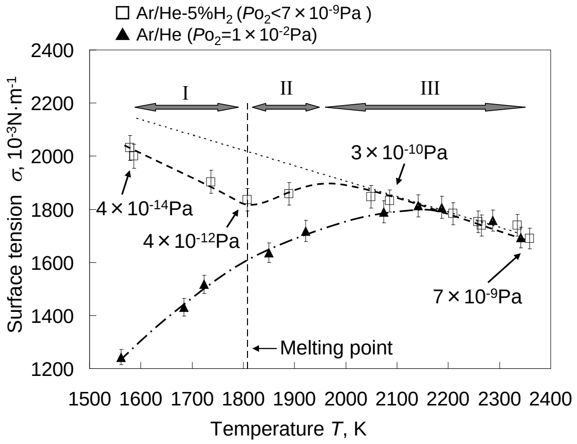

Fig. 2 surface tension of molten iron

When the sample temperature rises from 1580K to 1810K, the Po_2 of Ar/He-5%H_2 gas increased from 4×10^{-14}Pa to 4×10^{-12}Pa due to the chemical equilibrium of reaction (1). Since higher Po_2 normally induces a lower surface tension of molten metal due to oxygen adsorption at comparatively low temperature, surface tension becomes lower than that of the pure value as shown at the region I. What has to be noted is that the temperature elevation induces not only the increase of the Po_2 but also desorption of oxygen due to the temperature dependence of equilibrium constant for oxygen adsorption under H_2-containing gas atmosphere at the same time. The amount of oxygen adsorption is decided from the competition between these two mechanisms. When the oxygen adsorption becomes small due to high temperature even at high Po_2, the surface tension would approach the pure value as shown at the region II. When the equilibrium constant of oxygen adsorption becomes zero at high temperature, the surface tension of molten iron measured under the mixed gas of Ar/He-5%H_2 shows pure value at the region III.

Thus, the temperature dependence of surface tension of molten metals can not be described as a linear relationship against temperatures as long as pure surface of it does not appear when the Po_2 shows temperature dependence under reducing gas atmosphere such as H_2 and CO. The surface tension of molten metals should be described as functions of temperature and Po_2 as is the case for molten silver [4].

4. Summary

The surface tension of molten iron was measured by oscillating droplet method using EML under flow conditions of mixed gases of Ar/He and Ar/He-5%H_2. The surface tension was measured over wide temperature range of about 780K including undercooling condition. The “boomerang shape” temperature dependence of surface tension was experimentally confirmed at the Po_2 of 10^{-2}Pa. The temperature dependence of the surface tension showed kink at around 1850K under the Ar/He-5%H_2 atmosphere due to the competition between the temperature dependence of Po_2 and equilibrium constant of oxygen adsorption.

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