Melting and Flow Behavior of Fe-O Melts Heated by Plasma Arc

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In order to clarify the mechanism of change in molten pool geometry by plasma arc heating, the effects of oxygen contents of Fe-O melts on the geometry and flow behavior of molten pool were investigated. Also, the temperature coefficients of surface tension of Fe-O melts were examined in a temperature range from 1823 to 2023 K.

The depth of molten pool increased and the width of it decreased with increasing oxygen contents. Outward flow was observed at the surface of molten pool in the case of a low oxygen level and inward flow was observed in the case of a high oxygen level. The temperature coefficient of surface tension of Fe-O melts changed from a negative to positive value with increasing oxygen content. The ratio of depth to width of molten pool increased linearly with increasing temperature coefficient of surface tension. The geometry and flow behavior of molten pool were discussed in relation to the change of the temperature coefficient of surface tension with oxygen.

It was concluded from the experimental results that the Marangoni flow induced by the temperature difference in molten pool predominantly affect the geometry of molten pool.

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I. Introduction

Mukai *et al.*⁽¹⁾ showed that a local corrosion of refractory was caused by the motion of slag-film induced by the Marangoni effect due to a concentration gradient of SiO₂ on the surface of film. They⁽²⁾ also clarified the Marangoni flow in molten salts due to a temperature gradient even in the field of gravity of the earth.

In addition, the Marangoni flow due to the temperature gradient has been presumed to change the geometry of molten pool in arc welding⁽³⁾⁻⁽⁹⁾ when a surface active element such as sulfur was added to the mother metal. On the other hand, change in shape of arc caused by minor elements such as sulfur, aluminum, alkaline and alkaline earth metals in mother metal has been speculated to be another factor which affects the geometry of molten pool⁽¹⁰⁾⁻⁽¹³⁾. At the present stage, the mechanism of change in molten pool geometry accompanied with an increase in the minor element such as sulfur in molten metal has not yet been substantiated. Therefore, in this work, the geometry and flow behavior of molten pools in the Fe-O alloys heated by plasma arc were investigated under the following main experimental conditions.

Oxygen was selected as a minor element which is surface active against liquid iron. Fe-O alloys, which contained the smallest amount of impurities except for the deoxidizing elements, were provided as metal specimens. A contamination of molten pool by an atmosphere was prevented by the use of a sealed chamber. Fixed cooling conditions were kept in all experiments by applying the same size specimens of solid Fe-O alloys and a constant

temperature and flow rate of cooling water.

In order to discuss the possibility of occurrence of surface tension driven flow in molten pool, the effects of oxygen content and temperature on surface tension of Fe-O melts were investigated using a sessile drop method.

II. Experimental

1. Heating experiments by plasma arc

(1) Experimental apparatus and specimens

A plasma-jet-button-furnace shown in Fig. 1 was used

Fig. 1 Assembly in the chamber for the heating experiment by plasma arc.

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for making a molten pool in iron specimen. Argon gas, whose purity was over 99.999%, was used as a working gas; its flow rate was 120 ml/s. The temperature and flow rate of cooling water were controlled to be 298 ± 0.5 K and 130 ml/s, respectively.

Iron specimens were prepared by melting electrolytic iron in an argon atmosphere. The electrolytic iron contained 0.0022 mass% carbon, 0.0034 mass% sulfur, 0.0015 mass% phosphorus, 0.0017 mass% silicon, 0.0001 mass% copper and 0.050 mass% oxygen. After casting, the specimens were machined to have smooth surfaces. The specimens containing oxygen about 0.04 mass% were made of the electrolytic iron alone. Aluminum or pre-melted iron saturated with carbon was melted together with the electrolytic iron in order to make specimens contain oxygen less than 0.04 mass%. The quantity of aluminum or carbon added was under 1%, 0.53% relative to the weight of electrolytic iron, respectively. Fe_2O_3 powders of 99.5% purity were added to the electrolytic iron for the specimens of a higher oxygen level more than 0.04 mass%. Oxygen analysis of the specimens was performed using a LECO TC-136 Oxygen Determinator. Oxygen contents of the deoxidized specimens were analyzed paying careful attention not to detect oxygen in alumina clusters produced by deoxidation reaction; those contents ranged from 0.0006 to 0.0238 mass%.

Figure 2 shows the shape of a specimen. In the bottom of the specimen, three metal wires, whose composition was the same as the specimen, were inserted to detect the depth of molten pool. Further, other specimens were prepared to examine the flow behavior, inserting metal wires different in composition from the specimens or charging ZrO_2 powders in small holes on the surfaces of specimens.

(2) Experimental procedure

The specimen was attached to the water-cooled holder made of copper. Distance from plasma torch to surface of specimen was 10 mm. After evacuation, argon gas was introduced continuously. As soon as an arc was generated, the electrical output was raised to 25 V and 130 A. Then, heating by plasma arc was maintained for

Fig. 2 Cross-section of a specimen for the heating experiment by plasma arc.

70 or 90 s. During the heating, the shape of arc was photographed each 10 s and the direct observation of specimen surface was carried out. The specimen was allowed to cool down in the chamber after arc power was turned off. The width of solidified pool was measured by the use of a microscope and the specimen was cut along the metal wires to determine the depth of pool.

2. Surface tension measuring experiments

Surface tensions were measured in a temperature range from 1823 to 2023 K by means of the sessile drop method under an argon atmosphere. The scheme of the experimental apparatus is shown in Fig. 3.

The electrolytic iron, which was the same as the one used in the heating experiments by plasma arc, was prepared as iron samples for the surface tension measurements. For the experiments aiming at lower oxygen contents less than 0.005 mass%, iron samples were deoxidized at 1773 K in an argon-hydrogen gas mixture. With this treatment the oxygen contents of iron samples were reduced to about 0.005 mass%. Then, the measurements of surface tensions were performed in the argon-hydrogen gas mixture. For the experiments at higher oxygen contents, Fe_2O_3 powders were charged in a hole located at the top of iron sample before raising temperature. Experiments for examining the effect of the carbon on the surface tension of liquid iron were also conducted, charging high purity carbon powders in a hole of the iron sample.

Shapes of iron droplets on alumina or zirconia substrates were photographed at the prescribed temperatures. The values of surface tensions were determined by solving the Laplace equation from the photographed images of the iron droplets. This analysis was achieved by the use of a micro-computer, and its maximum error was about $\pm 2\%$. The experimental procedure and measurement of surface tension has been reported in detail elsewhere⁽¹⁴⁾.

Fig. 3 Furnace assembly for the surface tension measuring experiment.

III. Results

1. Relation between oxygen content in iron specimen and geometry of molten pool

Figure 4 shows the effects of oxygen content of the iron specimen on the geometries of molten pools, that is, depth D , width W and ratio D/W . Figure 4 indicates that the molten pool becomes narrower and deeper with increasing oxygen content of the specimen. The ratio D/W shows a steep increase up to about 0.01 mass% oxygen and then gradually increases in the range higher than 0.01 mass% oxygen. The heating time does not change the ratio D/W , which indicates that molten pool grows while maintaining a similar shape.

2. Effect of oxygen on surface tension of liquid iron

Figure 5 shows that surface tension decreases with increasing oxygen content at each temperature. However, the rate of surface tension reduction with oxygen content decreases with increasing temperature.

IV. Discussion

1. Shape of plasma arc

Ohji *et al.*⁽¹³⁾ have pointed out that an arc shrinks with an addition of surface active elements to metal specimen, and arc shrinking enhances the electromagnetic convection flow in molten pool, which increases the depth of molten pool.

Fig. 4 Effect of oxygen content of the specimen on the depth, width of molten pool and ratio of depth to width.

The shape of arc observed in this experiment was schematically shown in Fig. 6. Sizes of the narrow part, d_1 , and the wide part, d_2 , were measured from the photographed images of arc. d_1 and d_2 at 90 s of heating time are shown against oxygen content of the specimen in Fig. 6. Figure 6 shows that the shape of arc is not affected by the oxygen content of the specimen. d_1 and d_2 at 50 and 70 s of heating time were not affected by the oxygen content and were in good agreement with those at 90 s.

Fig. 5 Effect of oxygen content on the surface tension of liquid iron.

Fig. 6 Relation between oxygen content of the specimen and width of plasma arc at 90s.

It is reported that the aluminum addition to metal specimen accelerates the metal evaporation which may expand the arc and results in the formation of shallow molten pool⁽¹³⁾. In this work, the specimens containing oxygen below 0.04 mass% were deoxidized with aluminum. Figure 6 indicates that the presence of aluminum in metal specimens does not change the arc size. Moreover, in Fig. 4 no difference in the ratio D/W is found between the specimens deoxidized by aluminum and those deoxidized by carbon.

The above-mentioned experimental results indicate that arc size does not change in the concentration ranges of oxygen, aluminum and carbon in this work. Therefore, the change of molten pool geometry with oxygen contents of the specimens shown in Fig. 4 is not attributable to the change in arc shape.

2. Observation of flow behavior in molten pool

Figure 7 shows schematically cross-sections of the solidified specimens after the heating experiment by plasma arc. Metal wire markers different in composition from the metal specimen were combined with the specimens of a low oxygen level (Fig. 7(a)) and that of a high oxygen level (Fig. 7(b)). The metal wires inserted in the specimen of low oxygen content bend inward as shown in Fig. 7(a), which supports that an outward flow has occurred at the surface of molten pool. On the other hand, Fig. 7(b), the case of a high oxygen content (0.0750 mass%), supports the occurrence of inward flow. Al_2O_3 clusters were formed along the periphery of wire in molten pool by a reaction of the mother specimen and metal wires. The Al_2O_3 clusters prevent the wire from

dissolving into molten pool and then the metal wire can play a role of a marker to distinguish flow directions in molten pool.

Motions of ZrO_2 powders on the specimen surface were visually observed. The powders were drifting at the circumference of molten pool in the case of a low oxygen level, while in the case of a high oxygen level the powders moved at a high speed from the periphery to the center of molten pool and then they were drifting around the center. Each observation mentioned above substantiates the outward flow shown in Fig. 7(a) or the inward flow in Fig. 7(b), respectively.

Side views of molten pools were photographed immediately after turning off the arc power. Swelling up of the surface is observed at the center part of molten pool in the experiment using a specimen of a high oxygen level (0.0689 mass%) as shown in Fig. 8(b). The swelling up is presumed to be caused by a sink at the center of molten pool where molten iron is sucked in. This deformation of the surface due to the flow in molten pool is consistent with the flow characteristics of Marangoni convection⁽¹⁵⁾. On the other hand, the magnitude of swelling up of the surface in the case of a low oxygen level (0.0020 mass%) is very small as shown in Fig. 8(a). These phenomena also indicate that the flow conditions in molten pool would change with oxygen content of the specimen.

3. Interpretation of change of molten pool geometry by surface tension driven flow (Marangoni flow)

Heiple *et al.*⁽⁴⁾⁽⁵⁾ have proposed a process of the occurrence of surface tension driven flow in molten pool as follows:

In the process of arc welding, temperature is higher at the center of molten pool than at its circumference. When the temperature coefficient of surface tension of molten metal, $d\gamma/dT$, is negative, the surface tension at

Fig. 7 Profiles of metal wire markers in mother specimens after solidification in the case of combinations of (a) specimen of low oxygen level (0.0008 mass%) deoxidized by aluminum and markers of high oxygen level and (b) specimen of high oxygen level (0.075 mass%) and markers of low oxygen level (0.0007 mass%) deoxidized by the addition of 3 mass% aluminum.

Fig. 8 Side views of molten pool immediately after turning off arc power in the case using specimens of (a) low oxygen level (0.0020 mass%) and (b) high oxygen level (0.0689 mass%).

Fig. 9 Mechanism for penetration: (a) low level and (b) high content level of the surface active element in the specimen⁽⁴⁾⁽⁵⁾.

the circumference of molten pool, γ_A , is higher than that at the center part, γ_B , as shown in Fig. 9(a), which can be considered to generate an outward flow at the surface of molten pool. On the other hand, the positive value of $d\gamma/dT$, which may be realized with an addition of surface active elements as shown in Fig. 9(b), can cause the occurrence of inward flow.

However, Heiple *et al.*⁽⁴⁾⁽⁵⁾ did not measure the effects of surface active elements on the temperature coefficient of surface tension of the same molten metal that they used in the experiments for investigating the geometry of molten pool heated by plasma arc. Then, in this work, surface tension measurements of liquid Fe-O alloys prepared from the same electrolytic iron that used for the specimens in the heating experiments by plasma arc were conducted in order to examine the relation between the flow behavior of molten pool and the Marangoni flow proposed by Heiple *et al.*

Belton⁽¹⁶⁾ has derived eq. (1) by combining the Gibbs adsorption isothermal and the Langmuir adsorption isotherm:

$$\gamma^P - \gamma = R \cdot T \cdot \Gamma_i \cdot \ln(1 + K \cdot a_i) \quad (1)$$

where

- γ^P : surface tension of pure liquid iron,
- γ : surface tension of liquid iron when activity of surface active element i is equal to a_i ,
- R : gas constant,
- T : absolute temperature,
- Γ_i : saturated excess quantity for element i at liquid iron surface,
- K : constant for adsorption reaction.

Applying eq. (1) to Fe-O melts, eqs. (2)–(6) were obtained from the data shown in Fig. 5.

$$\gamma = 1.97 - 0.318 \cdot \ln(1 + 200 \cdot a_O) \text{ N/m at } 1823 \text{ K} \quad (2)$$

$$\gamma = 1.90 - 0.327 \cdot \ln(1 + 96 \cdot a_O) \text{ N/m at } 1873 \text{ K} \quad (3)$$

$$\gamma = 1.86 - 0.336 \cdot \ln(1 + 73 \cdot a_O) \text{ N/m at } 1923 \text{ K} \quad (4)$$

Fig. 10 Relation between oxygen content of liquid iron and the surface tension at various temperatures.

$$\gamma = 1.81 - 0.344 \cdot \ln(1 + 40 \cdot a_O) \text{ N/m at } 1973 \text{ K} \quad (5)$$

$$\gamma = 1.79 - 0.353 \cdot \ln(1 + 29 \cdot a_O) \text{ N/m at } 2023 \text{ K} \quad (6)$$

The procedure determining the values of γ^P , Γ_O , K and a_O has already been described in previous work⁽¹⁷⁾. These relations between oxygen contents and surface tensions expressed with eqs. (2)–(6) are shown in Fig. 10.

Figure 10 clearly shows that the value of $d\gamma/dT$ is negative below about 0.007 mass% oxygen and is positive above 0.007 mass% oxygen in Fe-O melts. An outward flow can be considered to occur for the specimen of the lower oxygen content less than 0.007 mass% because the surface tension at the circumference of molten pool is higher than that at the center part, whereas an inward flow occurs for the specimen of the higher oxygen level, which is consistent with the observation of the flow behavior in the present work and also the proposal of Heiple *et al.*⁽⁴⁾⁽⁵⁾

The Marangoni number M_a expressed by eq. (7) indicates the intensity of surface tension driven flow in fluids⁽¹⁸⁾.

$$M_a = \frac{(d\gamma/dT) \cdot \Delta T \cdot L}{\alpha \cdot \mu} \quad (7)$$

It can be presumed from the present heating conditions by plasma arc that the temperature difference between the center part and the circumference of molten pool, ΔT , is almost the same in all of the experiments. $W/2$, which is taken as the representative length L , shows a rather small change compared to the absolute values of $W/2$ in the present oxygen concentration range as shown in Fig. 4. Although the precise data on thermal diffusivity α and viscosity μ of liquid iron have not been available, the values of α and μ can be assumed to be nearly constant for all the specimens used in this work. Then, the temperature coefficient of surface tension, $d\gamma/dT$, can be regarded as an sole index representing the intensity of surface tension driven flow. In Fig. 11, $d\gamma/dT$ evaluated from eqs. (2)–(6) for each iron specimen is plotted against the ratio D/W . Figure 11 shows that the ratio D/W increases linearly with increasing $d\gamma/dT$. That is to say, the ratio D/W can be predominantly affected by the

Fig. 11 Relation between temperature coefficient of surface tension and ratio of depth to width of molten pool.

intensity of surface tension driven flow.

The above discussions can lead to the conclusion that Marangoni flow predominantly participates in the geometry of molten pool.

Specimens of oxygen contents below 0.04 mass% in the investigation of molten pool geometry contain aluminum or carbon. Therefore, it is needed to take account of the effects of these elements on the surface tension driven flow in molten pool.

Temperature coefficients of surface tension in Fe-C melts in the region of carbon contents below 0.5 mass% were obtained in the present study to be from -0.3 to -0.5 mN/m·deg. The values were close to those obtained by Kawai *et al.*⁽¹⁹⁾ and also to the value in this work, -0.43 mN/m·deg, for the Fe-O melts with about 0.001 mass% oxygen. For Fe-5 mass%Al melt, Ayushina⁽²⁰⁾ obtained a value of -0.24 mN/m·deg, which is close to the values of pure liquid iron measured by other workers⁽²¹⁾. Therefore, aluminum and carbon in the iron specimens of this study can be considered to have a minor effect on the temperature coefficient of surface tension, which is consistent with the results that the ratio D/W is almost the same between the specimens deoxidized with aluminum and carbon and is influenced only by oxygen content of the specimen as shown in Fig. 4.

V. Conclusion

The melting and flow behavior of Fe-O melts heated by plasma arc was investigated. Experiments were carried out by controlling the principal experimental conditions such as surface active components and the other impurities in the specimen, experimental atmosphere, and heating and cooling conditions of specimens. The results obtained are as follows:

(1) Depth of molten pool D increased and width W decreased with increasing oxygen content of the specimens heated by plasma arc. Ratio D/W , which designated geometry of molten pool, increased with increasing oxygen content.

(2) It was confirmed from visual direct observations

of molten pool and also from shapes of metal wire markers in the solidified specimens that an outward flow occurred at the surface of molten pool in the case of a low oxygen level, whereas an inward flow did in the case of a high oxygen level.

(3) Surface tension measurements by the sessile drop method have clarified that the temperature coefficient of surface tension of Fe-O melts changes from a negative to positive value with an increasing oxygen content. The above experimental result (2) can be reasonably interpreted from the behavior of the temperature coefficient of surface tension.

(4) Ratio D/W increased linearly with an increasing temperature coefficient of surface tension. The change in ratio D/W with oxygen, mentioned above in the result (1), could be explained in terms of the intensity of Marangoni flow.

The above experimental results can conclude that the Marangoni flow induced by the temperature difference in molten pool predominantly affect the geometry of molten pool.

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