Mass Transfer between Gas and Molten Iron in the Induction Furnace Equipped with Flow Controller

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The rate of nitrogen absorption of liquid iron was measured in a high-frequency-induction furnace equipped with a flow controller which makes it possible to control flow intensity near the melt surface under constant temperatures. The controller is composed of a secondary coil which is installed in the gap between the crucible and the induction coil, and an external coil connected to the secondary coil outside the furnace. By changing the turn number of the external coil, the magnetic field and electromagnetic force can be changed in the vicinity of the secondary coil. The rate of nitrogen absorption in molten iron was measured in a high frequency induction furnace, 20kW, 30kHz. The mass transfer coefficient (k_L) at 1873K could be changed from 8×10^{-4} to 5×10^{-4} m/s by the flow controller. The numerical analysis on the fluid flow and mass transfer was also made. The two dimensional Navier-Stokes equations for laminar and turbulent conditions were solved numerically. The mass-transfer coefficients were then obtained by solving the two dimensional nitrogen transfer equation for laminar flow, and by the eddy-cell model based on the computed values of energy dissipation rate for turbulent flow. A good agreement between estimated and observed values was obtained for turbulent flow condition.

Keywords: induction furnace, fluid flow, flow control, electromagnetic stirring, molten iron, mass transfer, nitrogen absorption, numerical analysis, k - ϵ model

I. Introduction

Laboratory-scale-induction furnaces have been used in numerous kinetic studies on metallurgical reactions such as gas-absorption or degasification reactions of molten iron⁽¹⁾⁻⁽³⁾. In these studies, reaction gas is introduced through a nozzle to the surface of liquid iron stirred inductively in a crucible (crucible-type reactor). To analyse the chemical reaction rate, it is important to know the effect of mass transfer included in measured rates. As the rate of mass transfer changes by the flow intensity and has weak dependence on temperature, it is desirable to change the flow intensity widely under a constant temperature in order to know the effect of mass transfer clearly. In the conventional crucible-type reactor, it is difficult to change the melt flow under a constant melt temperature, because the flow and temperature are both changed by the high frequency current imposed on the induction coil.⁽⁴⁾⁽⁵⁾

In the previous studies,⁽⁶⁾⁽⁷⁾ the authors investigated the method for flow control by a magnetic shield or a secondary coil. In the present study, the secondary coil method has

been applied to a kinetic study of nitrogen absorption in molten iron in an induction furnace.

II. Experimental

1. Method of experiment

Figure 1 shows the schematic diagram of the induction furnace equipped with a secondary coil. A four-turn-water-cooled copper coil was located close to the free surface of the molten iron, and both ends of the coil were extended out of the furnace and connected with the external coil. By changing the turn number, N_e , of the external coil in the range from 0 to 23 turns, the electromagnetic forces acting in a molten iron near the free surface is thought to be controlled widely.

Two different experiments have been made, one is the measurement of magnetic flux density and another is the nitrogen-absorption experiment. In the first experiment, a small search coil was used for measuring axial component of magnetic flux density along the axis of the induction coil. The secondary coil was located at the center of the induction coil. In the nitrogen-absorption experiment, a 0.5kg Fe-1mass%C alloy was melted in a MgO crucible of 40mm ID and 100mm high under argon atmosphere. After the melt temperature reached 1873 ± 20 K, nitrogen gas was

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Fig.1. Schematic diagram of the induction furnace equipped with a secondary coil.

introduced through the nozzle and the change in nitrogen concentration in liquid iron with time was measured.

In both experiments, a molybdenum shield of 52mm ID, 30mm high and 3mm thick was used instead of the secondary coil for comparison. The furnace used in this study was operated by a 30kHz, 20kW generator.

2. Results and discussion

2.1 Magnetic flux density

Figures 2(a) and 2(b) show the axial distribution of magnetic flux density inside the secondary coil and the molybdenum shield. As seen in Fig.2(a), the axial component of magnetic flux density, B_z , is suppressed by the secondary coil with decreasing turn number of the external coil. This is due to the fact that the induced current in the secondary coil has the opposite phase to that of the primary coil (work coil) and that it increases with decreasing the inductance of the secondary coil system. The calculated values of B_z shown in the same figure were obtained by the Biot-Savart's equation in which the measured currents in work coil and secondary coil were substituted.

In Fig.2(b), B_z is also suppressed in the molybdenum shield and the extent of suppression is greater than the case of the secondary coil. However, the extent of the suppression of magnetic field can not be varied by the shield method.

2.2 Absorption rate of nitrogen (previous results)

In the previous studies including our study⁽³⁾, the absorption rates of nitrogen in molten iron were satisfactorily described by the following first order rate equation:

$$\frac{d[N]}{dt} = \frac{A}{V} k_A ([N]_e - [N]) \quad (1)$$

where k_A is apparent rate constant, A surface area, V volume of iron, $[N]$ nitrogen concentration in the liquid bulk, $[N]_e$ equilibrium nitrogen concentration at the liquid surface. Many researchers pointed out that the value of k_A decreased

Fig.2. Axial distribution of B_z in the induction furnace with a secondary coil (a), and a molybdenum shield (b).

drastically by a small content of oxygen in liquid iron because of its strong surface-active nature. An example obtained by the present authors' group⁽³⁾ is given in Fig.3. In that study, effects of oxygen and carbon on k_A were investigated under two different furnace frequencies, 9.1 and 480kHz. In the case where carbon is added to liquid iron, which corresponds to oxygen free condition, the value of k_A is independent of carbon content and changes with the furnace frequency, f . As the stirring intensity is stronger for lower frequencies, the value of k_A at $f=9.1$ kHz is larger than that at $f=480$ kHz. When oxygen is added instead of carbon, the value of k_A decreases immediately and reaches a low value which is independent of f . These results indicate that the nitrogen absorption rate is mostly limited by the mass-transfer rate in the case of low oxygen contents, and by the chemical reaction rate in the case of high oxygen contents. Based on this prediction, Eq. (2) was derived:

$$\frac{1}{k_A} = \frac{1}{k_R(1-\theta_O)} + \frac{1}{k_L} \quad (2)$$

where k_R is reaction-rate constant, k_L mass-transfer coefficient, θ_O fraction of surface site of adsorbed oxygen. The results of k_L , k_R and θ_O obtained by applying observed values of k_A at 1873K to Eq.(2) are shown in Table 1.

Fig.3. Effects of furnace frequency on the apparent rate constant of nitrogen absorption in molten iron at various values of $[C]$ and $[O]$, which were obtained in the previous study⁽³⁾.

Table 1. Values of k_L , k_R and θ_0 .

	f=9.1kHz	f=480kHz
k_L/ms^{-1}	5.0×10^{-4}	3.3×10^{-4}
k_R/ms^{-1}	0.24×10^{-2}	
$[O]/K_O = \theta_O/(1-\theta_O)$; $K_O = 0.02-0.019\theta_O$		

2.3 Absorption rate of nitrogen (present results)

Equation (1) was well adopted to the present results, and values of k_A were determined. Substituting these values in Eq.(2), values of k_L were derived. The results obtained are given in Fig.4 in which the relation between k_L and the turn number of secondary coil is shown. As seen in the figure, k_L gives the smallest value at $Ne=0$, where the strongest flow suppression is expected. This value coincides with the value obtained for the molybdenum shield which gives stronger suppression of magnetic flux density than the secondary coil as shown in Fig.2. As the turn number increases, the value of k_L increases gradually and reaches a maximum in the case without flow control.

There can be seen 1.6 fold change in the value of k_L by using the secondary coil method. As seen in Table 1, this ratio is comparable to that obtained in the previous study (1.5) which has been realized by frequency change. To install the secondary coil to a furnace must be easier and more advantageous than to change furnace frequency. By the use of this method, the kinetic study on various metallurgical reactions can be substantially developed. If this method is applied to the industrial furnace, local erosion of furnace refractory may be reduced.

Fig.4. Observed values of k_L under flow control by secondary coil and molybdenum shield.

III. Calculations

1. Method of calculation

1.1 Fluid flow

Assumptions made in the analysis are as follows:

- (1) The electromagnetic field is not affected by the fluid flow.
- (2) The temperature of liquid iron is uniform.
- (3) The system is axisymmetric and in steady state.
- (4) The free surface of liquid iron is flat.

Assumption (1) allows us to solve the electromagnetic field independently of the flow field. The mutual inductance method⁽⁸⁾ was applied to obtain the induced current, J , and magnetic flux densities, B_r and B_z , in liquid iron. The time-averaged electromagnetic forces were calculated by the following equation:

$$f_r = \frac{1}{2} \text{Re}\{J \cdot B_z^*\}; \quad f_z = -\frac{1}{2} \text{Re}\{J \cdot B_r^*\} \quad (3)$$

In this study, numerical calculation was made for both laminar flow and turbulent flow. The governing equations for turbulent flow are expressed by Eqs.(4) to (8) in the coordinate system shown in Fig.5.

Fig.5. Coordinate system for numerical analysis.

$$\frac{1}{r} \frac{\partial u}{\partial r} + \frac{\partial v}{\partial z} = 0 \quad (4)$$

$$u \frac{\partial u}{\partial r} + v \frac{\partial u}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial r} + \frac{1}{r} \frac{\partial}{\partial r} \left(2\nu_e r \frac{\partial u}{\partial r} \right) - \frac{2}{r^2} \nu_e u + \frac{\partial}{\partial z} \left(\nu_e \left(\frac{\partial v}{\partial r} + \frac{\partial u}{\partial z} \right) \right) + \frac{1}{\rho} f_r \quad (5)$$

$$u \frac{\partial v}{\partial r} + v \frac{\partial v}{\partial z} = -\frac{1}{\rho} \frac{\partial p}{\partial z} + \frac{1}{r} \frac{\partial}{\partial r} \left(\nu_e r \left(\frac{\partial v}{\partial r} + \frac{\partial u}{\partial z} \right) \right) + \frac{\partial}{\partial z} \left(2\nu_e \frac{\partial v}{\partial z} \right) - g + \frac{1}{\rho} f_z \quad (6)$$

$$u \frac{\partial k}{\partial r} + v \frac{\partial k}{\partial z} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \nu_e \frac{\partial k}{\partial r} \right) + \frac{\partial}{\partial z} \left(\nu_e \frac{\partial k}{\partial z} \right) + G - \varepsilon \quad (7)$$

$$u \frac{\partial \varepsilon}{\partial r} + v \frac{\partial \varepsilon}{\partial z} = \frac{1}{r} \frac{\partial}{\partial r} \left(r \nu_e \frac{\partial \varepsilon}{\partial r} \right) + \frac{\partial}{\partial z} \left(\nu_e \frac{\partial \varepsilon}{\partial z} \right) + \frac{C_1}{k} \varepsilon G - \frac{C_2}{k} \varepsilon^2 \quad (8)$$

where ν_e is the effective kinematic viscosity given by the sum of the kinematic viscosity of liquid iron and turbulent kinematic viscosity as follows:

$$\nu_e = \nu + \nu_t; \quad \nu_t = C_D k^2 / \varepsilon \quad (9)$$

k and ε are turbulence energy and energy-dissipation rate, respectively. The generation term of turbulence, G , in Eqs. (7) and (8) is given by

$$G = \nu_t \left[2 \left(\left(\frac{\partial v}{\partial z} \right)^2 + \left(\frac{\partial u}{\partial r} \right)^2 + \left(\frac{u}{r} \right)^2 \right) + \left(\frac{\partial v}{\partial r} + \frac{\partial u}{\partial z} \right)^2 \right] \quad (10)$$

The constants included in the equations are as follows:

$$C_D = 0.09, C_1 = 1.44, C_2 = 1.92, \sigma_k = 1, \sigma_\varepsilon = 1.3$$

The boundary conditions were given by

$$\begin{aligned} r = 0, 0 < z < z_1 : u = \frac{\partial v}{\partial r} = \frac{\partial k}{\partial r} = \frac{\partial \varepsilon}{\partial r} &= 0 \\ r = r_1, 0 < z < z_1 : u = v = k = \varepsilon &= 0 \\ z = 0, 0 < r < r_1 : u = v = k = \varepsilon &= 0 \\ z = z_1, 0 < r < r_1 : \frac{\partial u}{\partial z} = v = \frac{\partial k}{\partial z} = \frac{\partial \varepsilon}{\partial z} &= 0 \end{aligned} \quad (11)$$

Wall function was also used for the velocity profile near the wall.

The equations depicted above were transformed into dimensionless form and modified by the stream function, ψ , and vorticity, ζ . The upwind-difference scheme⁽⁹⁾ and the successive-over-relaxation technique⁽¹⁰⁾ were adopted for solving the equations. The number of grid points for iterative computation was 21×61 for r and z direction. For the condition of convergence in the computation, $|\Delta \zeta / \zeta|_{\max} < 10^{-4}$ was employed.

In the case of laminar flow, Eqs. (4) to (6) with constant kinematic viscosity were solved by the same method depicted above.

1.2 Mass transfer

For laminar flow, Eqs. (12) and (13) were solved by using upwind-difference scheme and ADI method⁽¹¹⁾.

$$\frac{\partial C}{\partial t} + u \frac{\partial C}{\partial r} + v \frac{\partial C}{\partial z} = D \left[\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C}{\partial r} \right) + \frac{\partial^2 C}{\partial z^2} \right] \quad (12)$$

$$\begin{aligned} t = 0 : C &= C_0 \\ r = 0, 0 < z < z_1 : \frac{\partial C}{\partial r} &= 0 \\ r = r_1, 0 < z < z_1 : \frac{\partial C}{\partial r} &= 0 \\ z = 0, 0 < r < r_1 : \frac{\partial C}{\partial z} &= 0 \\ z = z_1, 0 < r < r_1 : C &= C_e \end{aligned} \quad (13)$$

The mass transfer coefficient was calculated from the concentration gradient near the free surface.

In the case of turbulent flow, a turbulent mass transfer model was applied for the estimation of mass-transfer coefficient. The model used is the eddy-cell model proposed by Lamont and Scott⁽¹²⁾, which has been developed from Danckwerts' surface-renewal theory. The surface renewal by turbulent eddies with various sizes was assumed in their model. The mass-transfer coefficient is given by the following equation:

$$k_L = \alpha D^{1/2} (\varepsilon / \nu)^{1/4} \quad (14)$$

where α is an unknown constant. Kataoka and Miyauchi⁽¹³⁾ derived the same equation as Eq. (14) through a different way. They assumed that the surface renewal was made only by the smallest eddy. They found that in the case $\alpha = 0.5$, Eq. (14) agreed well with the observed k_L for the absorption of CO_2 from the surface of turbulent water tunnel.

To estimate the value of k_L by Eq. (14), the calculated values of ε were averaged along the surface and substituted in Eq. (14).

2. Results and discussion

Figure 6 shows the calculated magnetic fields for three cases, (a) without flow control, (b) with secondary coil ($\text{Ne} = 0$), and (c) with molybdenum shield. It is seen from the figures that the magnetic field lines are distorted in the vicinity of the secondary coil and the shield.

Flow patterns of molten iron are indicated in Fig. 7 for turbulent flow condition. There can be seen four recirculating flows with different flow direction to each other in the case without flow control. As Ne decreases, the upper two flows become weak and finally disappear and one large clockwise flow occupies the greater part of the flow area. Similar results has been reported by Vives and Ricou⁽¹⁴⁾ in their study of the screen in the induction furnace.

The distributions of the effective kinematic viscosity (ν_e / ν) are shown in Fig. 8. In all cases, values of ν_e / ν show wide distribution and give maximum at the center of the free surface. Its value is found to increase with decreasing Ne .

Finally, the estimated value of k_L by Eq. (14) are compared with the observed results in Fig. 9. Estimated values assuming laminar flow are far less than the observed results and they fail to indicate the effect of Ne as seen in

Fig.6. Calculated magnetic fields.

the figure. On the contrary, the estimated values by Eq.(14) give a satisfactory agreement with the observed results.

IV. Conclusions

The secondary coil method which enables local flow control under a constant temperature has been applied to a laboratory-scale-induction furnace, and the nitrogen-absorption rate in molten iron has been measured under various flow intensities. The following results have been

Fig.7. Computed stream line patterns for turbulent flow condition at various Ne.

obtained.

1. The magnetic flux densities in the furnace could be suppressed by the secondary coil as well as by the molybdenum shield.
2. The mass transfer coefficients for nitrogen absorption could be reduced from 8×10^{-4} to 5×10^{-4} m/s with the secondary coil method.
3. The effects of the secondary coil on the electromagnetic field and flow field have been estimated by numerical simulations.
4. The mass-transfer coefficient calculated by the eddy-cell model agreed well with the observed values.

Fig.8. Computed distribution of effective kinematic viscosity (v_e/v) at various Ne .

Fig.9. Comparison between observed and calculated values of k_L .

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