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Thermodynamic Analysis on the Minimum of Oxygen Content in the Deoxidation Equilibrium Curve in Liquid Iron

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Abstract: The minimum of oxygen content in the deoxidation equilibrium in liquid iron was thermodynamically analyzed in the present paper. Two criteria were developed to determine the existence of the minimum. The first criterion was $0 \le x\gamma_M^M + y\gamma_O^M \le \min\left(x\big/4.606[\%M]_{ex}^2, (xe_M^M + ye_O^M)^2\big/3.474x\right)$ with $xe_M^M + ye_O^M < 0$, or $x\gamma_M^M + y\gamma_O^M < 0$. And the second criterion was $(xe_M^O + ye_O^O) + y\big/2.303[\%O]_{ex} > 0$. The criteria in terms of first-order activity interaction parameters were the special case of present thermodynamic analysis with neglecting the second-order activity interaction parameters. They were not fit for the case of $xe_M^M + ye_O^M > 0$, in which case the criteria in terms of second-order activity interaction parameters should be taken into account to determine the existence of the minimum. The value 0.11 of e_{Si}^{Si} was smaller based on the existence of the minimum for the Fe-O-Si system. It was guaranteed that the minimum value of oxygen content on the deoxidation equilibrium curve existed at silicon content 20 mass%, when the value 0.32 of e_{Si}^{Si} was chosen, and the second-order activity interaction coefficients γ_{Si}^{Si} and γ_O^{Si} satisfied the condition $\gamma_{Si}^{Si} + 2\gamma_O^{Si} = -1.54 \times 10^{-3}$.

Keywords: deoxidation equilibrium curve, the minimum of oxygen content, first-order activity interaction coefficients, second-order activity interaction coefficients

Introduction

The deoxidation equilibrium curve can be obtained depending on the calculation of the deoxidation equilibrium in liquid steel by using thermodynamic data for different elements, as shown in Figure 1 [1, 2]. According to Figure 1, there are minima for the Fe-O-Al, Fe-O-C and Fe-O-Cr systems, but no minimum exists in the deoxidation equilibrium curve of Fe-O-Si system.

Silicon–oxygen equilibrium in liquid iron was experimented by Gokcen and Chipman in the early 1950s [3]. Their results showed that there was no minimum for the Fe-O-Si system, even the silicon content up to 15 mass%. However, it was reported recently by Shibaev et al. [4] that the minimum occurred at 20 mass% Si, who measured the solubility of oxygen in iron–silicon melts in equilibrium with silica at 1,873 K, as shown in Figure 2. The criteria for a minimum developed by Hone et al. [1] and Wei [2] are not fit for the condition that the deoxidizer content is fairly high. The aim of the present study is to correct the criteria for the existence of the minimum of oxygen content in the deoxidation equilibrium curve.

Thermodynamic analysis

The reaction between deoxidizer and oxygen can be represented as follows:

$$x[M] + y[O] = M_xO_y \tag{1}$$

$$m = a_M^x a_O^y \tag{2}$$

where x and y are stoichiometric coefficients, [M] and [O] are metallic deoxidizer and oxygen in liquid iron, respectively, m is the reciprocal of equilibrium constant, a_M and a_O are the activities of [M] and [O]. By replacing a_M and a_O with $f_M[\%M]$ and $f_O[\%O]$, respectively, eq. (3) is obtained.

$$\lg m = x \lg f_M + x \lg[\%M] + y \lg f_O + y \log[\%O] \tag{3}$$

The dependence of f_M and f_O on composition can be expressed as eqs (4) and (5) [5].

$$\lg f_M = e_M^M[\%M] + e_M^O[\%O] + \gamma_M^M[\%M]^2 + \gamma_M^O[\%O]^2 + \gamma_M^{M,O}[\%M][\%O] \tag{4}$$

$$\lg f_O = e_O^O[\%O] + e_O^M[\%M] + \gamma_O^O[\%O]^2 + \gamma_O^M[\%M]^2 + \gamma_O^{M,O}[\%M][\%O] \tag{5}$$

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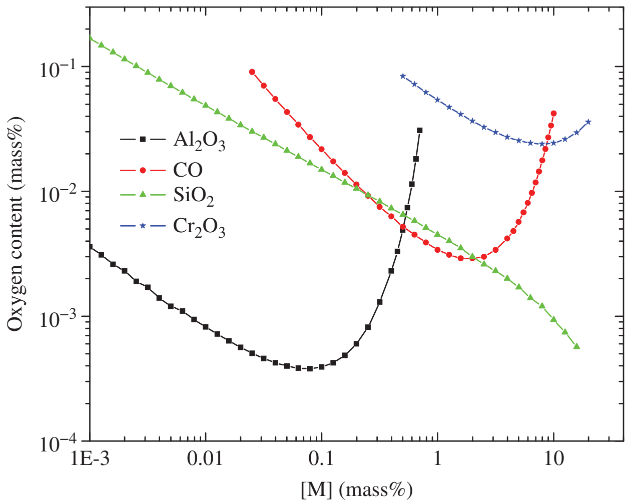

Figure 1: Deoxidation equilibrium curve of different elements in iron melt at 1,873 K.

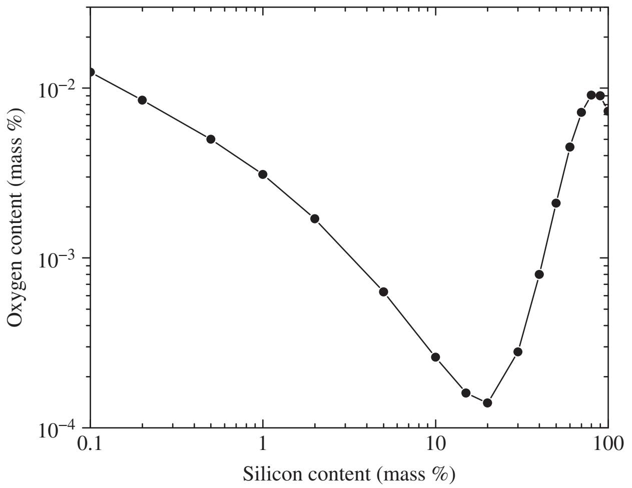

Figure 2: Oxygen solubility in Fe–Si melts in equilibrium with silica at 1,873 K.

Neglecting the higher order terms of [%O] and the second-order terms $\gamma_M^{M,O}[\%M][\%O]$ and $\gamma_O^{M,O}[\%M][\%O]$, eqs (4) and (5) reduce to eqs (6) and (7), respectively.

$$\lg f_M = e_M^M[\%M] + e_M^O[\%O] + \gamma_M^M[\%M]^2 \tag{6}$$

$$\lg f_O = e_O^O[\%O] + e_O^M[\%M] + \gamma_O^M[\%M]^2 \tag{7}$$

Substitution of eqs (6) and (7) into eq. (3) yields

$$\begin{aligned}\lg m = {} & [\%M](xe_M^M + ye_O^M) + [\%O](xe_M^O + ye_O^O) \\ & + [\%M]^2(x\gamma_M^M + y\gamma_O^M) + x\log[\%M] + y\log[\%O]\end{aligned} \tag{8}$$

If there is a minimum, the [%O] versus [%M] curve should meet the following two conditions:

$$\frac{d[\%O]}{d[\%M]} = 0 \tag{9}$$

$$\frac{d^2[\%O]}{d[\%M]^2} > 0 \tag{10}$$

Equation (11) is obtained by differentiating eq. (8) and further rearranging:

$$\frac{d[\%O]}{d[\%M]} = -\frac{xe_M^M + ye_O^M + 2[\%M](x\gamma_M^M + y\gamma_O^M) + \frac{x}{2.303[\%M]}}{xe_M^O + ye_O^O + \frac{y}{2.303[\%O]}} = 0 \tag{11}$$

With the first derivative zero, the second derivative of eq. (8) is

$$\frac{d^2[\%O]}{d[\%M]^2} = \frac{-2(x\gamma_M^M + \gamma r_O^M) + \frac{x}{2.303[\%M]^2}}{xe_M^O + ye_O^O + \frac{y}{2.303[\%O]}} > 0 \tag{12}$$

According to eq. (11), eq. (13) is obtained.

$$xe_M^M + ye_O^M + 2[\%M](x\gamma_M^M + y\gamma_O^M) + \frac{x}{2.303[\%M]} = 0 \tag{13}$$

If $x\gamma_M^M + y\gamma_O^M = 0$, eq. (13) is a linear equation, to ensure the existence of a positive real solution for it, the following condition should be satisfied:

$$xe_M^M + ye_O^M < 0 \tag{14}$$

However, eq. (13) is a quadratic equation if $x\gamma_M^M + y\gamma_O^M \neq 0$. Equation (15) meets the condition that the quadratic equation has real solutions. And the corresponding real solutions are given in eq. (16).

$$(xe_M^M + ye_O^M)^2 \geq 3.474x(x\gamma_M^M + y\gamma_O^M) \tag{15}$$

$$[\%M] = \frac{-(xe_M^M + ye_O^M) \pm \sqrt{(xe_M^M + ye_O^M)^2 - 3.474x(x\gamma_M^M + y\gamma_O^M)}}{4(x\gamma_M^M + y\gamma_O^M)} \tag{16}$$

For the case of $x\gamma_M^M + y\gamma_O^M < 0$, there is always a positive real solution for eq. (13) no matter whether $xe_M^M + ye_O^M$ is positive or negative. And eq. (15) is always satisfied if $x\gamma_M^M + y\gamma_O^M < 0$.

But when it comes to the case of $x\gamma_M^M + y\gamma_O^M > 0$, eq. (14) should be satisfied to ensure the existence of positive real solutions for eq. (13). In this case, there are two positive real solutions. This is similar to the work of St. Pierre [6], but in which the constraints were not discussed such as $x\gamma_M^M + y\gamma_O^M > 0$ and the following eqs (18) and (19).

Equation (12) is needed to determine whether the positive real solution of eq. (13) is the minimum in the deoxidation equilibrium curve. Let $[\%\mathrm{M}]_{\mathrm{ex}}$ be the positive real solution of eq. (13) and $[\%\mathrm{O}]_{\mathrm{ex}}$ be the corresponding oxygen content obtained by substitution of $[\%\mathrm{M}]_{\mathrm{ex}}$ into eq. (8). Hence, eq. (12) can be written as

$$\frac{-2(x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}})+\frac{x}{2.303[\%\mathrm{M}]_{\mathrm{ex}}^{2}}}{xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}}+\frac{y}{2.303[\%\mathrm{O}]_{\mathrm{ex}}}}>0 \tag{17}$$

For the case of $x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}}\leq 0$, since the denominator term of eq. (11) is positive, the denominator term of eq. (17) follows that

$$(xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}})+y/2.303[\%\mathrm{O}]_{\mathrm{ex}}>0 \tag{18}$$

For the case of $x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}}\leq 0$, it is reasonable that the smaller solution $\frac{-(xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}})-\sqrt{(xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}})^{2}-3.474x(x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}})}}{4(x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}})}$ of eq. (13) should be the minimum, since the oxygen in liquid iron decreases initially as the content of deoxidizer increases. As a consequence, the denominator term of eq. (11) should be positive. It means that it is also positive for the denominator term of eq. (17). Therefore the denominator term of eq. (11) should be greater than zero and satisfy the following equation:

$$x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}}<x\Big/4.606[\%\mathrm{M}]_{\mathrm{ex}}^{2} \tag{19}$$

From the thermodynamic analysis above, two criteria are developed for determining the existence of the minimum in the deoxidation equilibrium curve.

The first criterion is $0\leq x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}}\leq \min\left(x\Big/4.606[\%\mathrm{M}]_{\mathrm{ex}}^{2},\ (xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}})^{2}\Big/3.474x\right)$ with $xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}}<0$, or $x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}}<0$.

And the second criterion is $(xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}})+y/2.303[\%\mathrm{O}]_{\mathrm{ex}}>0$ where $\min(A,B)$ is the minimum between A and B. It should be noted that there is always a minimum no matter whether $xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}}$ is positive or negative, in the case of $x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}}<0$ and $(xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}})+y/2.303[\%\mathrm{O}]_{\mathrm{ex}}>0$.

Discussion

The limitation of the criteria for the minimum in terms of first-order activity interaction parameters

Since the amount of deoxidizer addition is not large in steelmaking practice in general, the first-order activity interaction parameters are adopted to determine the existence of the minimum in the deoxidation equilibrium curve [1, 2, 7, 8]. In other words, $\gamma_{\mathrm{M}}^{\mathrm{M}}$ and $\gamma_{\mathrm{O}}^{\mathrm{M}}$ are both regarded as zero. The first criterion for the minimum is $xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}}<0$ in this condition, which is consistent with eq. (14). It can be noted that this criterion is the special case of $x\gamma_{\mathrm{M}}^{\mathrm{M}}+y\gamma_{\mathrm{O}}^{\mathrm{M}}=0$ in Section 2.

In the work of Wei [2], the second criterion is given as

$$xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}}<0 \tag{20}$$

It is considered that eq. (20) is questionable. Of course, $xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}}>0$ satisfies eq. (18). On the other hand, it is not guaranteed that the value of $-(xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}})$ is smaller than that of $y/2.303[\%\mathrm{O}]_{\mathrm{ex}}$ in the case of $xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}}<0$. Thus, the second criterion is eq. (18) rather than eq. (20). $[\%\mathrm{O}]_{\mathrm{ex}}$ is obtained by eqs (21) and (22) in terms of first-order criterion parameters. Then, the existence of the minimum is confirmed by substitution of $[\%\mathrm{O}]_{\mathrm{ex}}$ into eq. (18).

$$[\%\mathrm{M}]_{\mathrm{ex}}=-\frac{x}{2.303(xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}})} \tag{21}$$

$$\begin{aligned}\lg m=[\%\mathrm{M}]_{\mathrm{ex}}(xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}})+[\%\mathrm{O}]_{\mathrm{ex}}(xe_{\mathrm{M}}^{\mathrm{O}}+ye_{\mathrm{O}}^{\mathrm{O}})\\+x\log[\%\mathrm{M}]_{\mathrm{ex}}+y\log[\%\mathrm{O}]_{\mathrm{ex}}\end{aligned} \tag{22}$$

It is worth noting that only the first criterion is not sufficient for the existence of the minimum. It is not persuasive enough to confirm the existence of a minimum in the work of St. Pierre and Blackburn [7] and the work of Feldman and Kirkaldy [8].

The value of activity interaction parameter can be judged by the existence of the minimum in the deoxidation equilibrium curve. It can be concluded that the value −0.13 of $e_{\mathrm{O}}^{\mathrm{C}}$ is incorrect [2], since there is no minimum for the Fe-O-C system with $e_{\mathrm{O}}^{\mathrm{C}}=-0.13$. This is the contribution of the criteria in terms of the first-order activity interaction parameters.

However, there are limitations of criteria for the minimum in terms of first-order activity interaction parameters.

First, it is only suitable to the range that $\lg f_{\mathrm{M}}^{\mathrm{M}}$ and $\lg f_{\mathrm{O}}^{\mathrm{M}}$ have a linear relationship with [%M] due to the neglect of second-order interaction parameters. As [%M] increases, it is also lager for the degree of $\lg f_{\mathrm{M}}^{\mathrm{M}}$ and $\lg f_{\mathrm{O}}^{\mathrm{M}}$ deviating from the linear relationship with [%M]. The influence of Ti elements on the oxygen solubility in liquid iron at 1,873 K is shown in Figure 3 [8], in which the full line is the theoretical solubility curves calculated in terms of first-order activity interaction parameters, and the dots are experimental data from the work of Fruehan [9]. It can be seen from Figure 3 that the position of the minimum is accurately predicted by the criteria in terms of the first-order activity interaction parameters for some elements. But the deviation between the calculated value and the experimental value is larger as [%M] increases.

Second, it is concluded that there is no minimum in the case of $xe_{\mathrm{M}}^{\mathrm{M}}+ye_{\mathrm{O}}^{\mathrm{M}}>0$ in Refs. [1] and [2]. Thus the

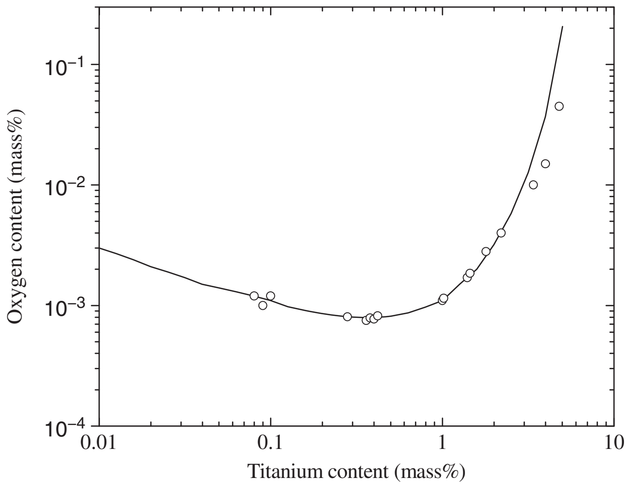

Figure 3: Influence of Ti element on the oxygen solubility in liquid iron at 1,873 K (reproduced from Ref. [8]).

minimum is considered to be inexistence for the Fe-O-Si system. According to Figure 2, there is a minimum in the deoxidation equilibrium curve at high silicon content. Thus, even if $xe_M^M + ye_O^M > 0$, there is still a minimum when it satisfies the condition of $x\gamma_M^M + y\gamma_O^M < 0$ and $(xe_M^O + ye_O^O) + y/2.303[\%O]_{ex} > 0$.

The deoxidation equilibrium curve of silicon in liquid iron

There is a disagreement in the literature on the existence of the minimum in the deoxidation equilibrium curve for Fe-O-Si system. With the criteria in terms of first-order activity interaction parameters, there is a minimum with the data from Refs. [10] and [11], but not with data from Refs. [1] and [2]. The data used on calculation and the results are shown in Tables 1 and 2, respectively. The values of $[\%Si]_{ex}$ and $[\%O]_{ex}$ in Table 2 are obtained by eqs (21) and (22), respectively.

Table 1: Data on calculation of silicon deoxidation equilibrium curve at 1,873 K.

x	y	e_{Si}^{Si}	e_O^{Si}	e_{Si}^O	e_O^O	lg m	Note
1	2	0.32	−0.14	−0.25	−0.20	−4.66	Refs [1, 2]
		0.11	−0.131	−0.23	−0.20	−4.64	Refs [10, 11]

The difference of results in Table 2 is mainly caused by the value of e_{Si}^{Si}. There is a big disagreement about activity interaction parameters obtained by different research, due to the experimental methods, experimental conditions, the high temperature control, etc.

There is a minimum at silicon content 2.86 mass% for the Fe-O-Si system with data from Refs [10] and [11]. It differs from the experimental results as shown in Figure 2. According to Figure 2, the minimum of oxygen content is 1.4 ppm at the silicon content 20 mass% [4].

Depending on the above thermodynamic analysis, the first criterion for a minimum is derived from eq. (13). Substitution of $[\%Si]_{ex} = 20$ and $e_{Si}^{Si} + 2e_O^{Si} = -0.152$ into eq. (13) yields

$$\gamma_{Si}^{Si} + 2\gamma_O^{Si} = 3.26 \times 10^{-3} \tag{23}$$

Then application of the second criterion, substitution of $e_{Si}^O + 2e_O^O = -0.63$, $[\%Si]_{ex} = 20$ and $[\%O]_{ex} = 1.4 \times 10^{-4}$ into eq. (17) yields

$$\gamma_{Si}^{Si} + 2\gamma_O^{Si} < 5.43 \times 10^{-4} \tag{24}$$

Obviously, the value of eq. (23) is out of the range of eq. (24). It can be concluded that the value 0.11 of e_{Si}^{Si} is smaller.

The data of $e_{Si}^{Si} = 0.32$ and $e_O^{Si} = -0.14$ from Refs. [1] and [2] and the effect of second-order activity interaction parameters are taken into account. Substitution of $[\%Si]_{ex} = 20$ and $e_{Si}^{Si} + 2e_O^{Si} = 0.04$ into eq. (13) yields

$$\gamma_{Si}^{Si} + 2\gamma_O^{Si} = -1.54 \times 10^{-3} \tag{25}$$

The second criterion is also satisfied by substituting $[\%O]_{ex} = 1.4 \times 10^{-4}$ and $e_{Si}^O + 2e_O^O = -0.65$ into eq. (18). It means that it determines the existence of the minimum at silicon content 20 mass% theoretically.

It is interesting to note that the values of γ_{Si}^{Si} and γ_O^{Si} are −0.0021 and 0, respectively, in Ref. [10]. Their sum is similar to the calculated result as shown in eq. (25) in the present paper.

Table 2: Verification on the minimum of Fe-O-Si system.

$e_{Si}^{Si} + 2e_O^{Si}$	$e_{Si}^O + 2e_O^O$	[%Si]/mass%	[%O]/ppm	$y/2.303[\%O]_{ex}$	The minimum
0.04	−0.65	–	–	–	No*
−0.152	−0.63	2.86	47	1.85 × 102	Yes**

*Data from Refs [1] and [2].
** Data from Refs [10] and [11].

Conclusions

Two criteria are developed for determining the existence of the minimum in the deoxidation equilibrium curve in liquid iron in the present paper. The first criterion is $0 \le x\gamma_{\mathrm{M}}^{\mathrm{M}} + y\gamma_{\mathrm{O}}^{\mathrm{M}} \le \min\left(x \big/ 4.606[\%\mathrm{M}]_{\mathrm{ex}}^2, \left(xe_{\mathrm{M}}^{\mathrm{M}} + ye_{\mathrm{O}}^{\mathrm{M}}\right)^2 \big/ 3.474x\right)$ with $xe_{\mathrm{M}}^{\mathrm{M}} + ye_{\mathrm{O}}^{\mathrm{M}} < 0$ or $x\gamma_{\mathrm{M}}^{\mathrm{M}} + y\gamma_{\mathrm{O}}^{\mathrm{M}} < 0$. And the second criterion is $\left(xe_{\mathrm{M}}^{\mathrm{O}} + ye_{\mathrm{O}}^{\mathrm{O}}\right) + y/2.303[\%\mathrm{O}]_{\mathrm{ex}} > 0$.

The criteria in terms of first-order activity interaction parameters are the special case of present thermodynamic analysis with neglecting the second-order activity interaction parameters. They are not fit for the case of $xe_{\mathrm{M}}^{\mathrm{M}} + ye_{\mathrm{O}}^{\mathrm{M}} > 0$. In this case, the criteria in terms of second-order activity interaction parameters are taken into account to determine the existence of the minimum.

The value 0.11 of $e_{\mathrm{Si}}^{\mathrm{Si}}$ is smaller based on the existence of the minimum in the deoxidation equilibrium curve for the Fe-O-Si system. It is guaranteed that the minimum value of oxygen content on the deoxidation equilibrium curve existed at silicon content 20 mass%, when the value 0.32 of $e_{\mathrm{Si}}^{\mathrm{Si}}$ is chosen, and the second-order activity interaction coefficients $\gamma_{\mathrm{Si}}^{\mathrm{Si}}$ and $\gamma_{\mathrm{O}}^{\mathrm{Si}}$ satisfy the condition $\gamma_{\mathrm{Si}}^{\mathrm{Si}} + 2\gamma_{\mathrm{O}}^{\mathrm{Si}} = -1.54 \times 10^{-3}$.

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List of symbols

γ_i^i	The second-order self-interaction parameters of element *i* in liquid iron on 1 mass% solution standard
γ_i^j	The second-order interaction parameter of element *j* on element *i* in liquid iron on 1 mass% solution standard
$\gamma_i^{i,j}$	The cross-product second-order interaction coefficient of elements *i* and *j* on element *i* in liquid iron on 1 mass% solution standard
a_i	Activity of element *i* in liquid iron
e_i^i	The first-order self-interaction parameters of element *i* in liquid iron on 1 mass% solution standard
e_i^j	The first-order interaction parameter of element *j* on element *i* in liquid iron on 1 mass% solution standard
f_i	Activity coefficient of element *i* on 1 mass% solution standard
f_i^j	The influence of element *j* on f_i on 1 mass% solution standard
$[i]$	Elements *i* in liquid iron
$[\%i]$	Weight percentage of element *i* in liquid iron
m	The reciprocal of equilibrium constant
$\min(A,B)$	The minimum between *A* and *B*
$\mathrm{M}_x\mathrm{O}_y$	Deoxidizing product
$[\%\mathrm{M}]_{\mathrm{ex}}$	The positive real solution of eq. (13)
$[\%\mathrm{O}]_{\mathrm{ex}}$	The corresponding oxygen content obtained by substitution of $[\%\mathrm{M}]_{\mathrm{ex}}$ into eq. (8)
x	Stoichiometric coefficient in deoxidizing product
y	Stoichiometric coefficient in deoxidizing product

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