Solubility of Chlorine in Aluminosilicate Slag Systems

Taro HIROSUMI and Kazuki MORITA1)

Formerly Graduate Student, Department of Metallurgy, The University of Tokyo. Now at Steel Research Laboratories, Nippon Steel Corporation, Shintomi, Futtsu, Chiba 293-8511 Japan. 1) Department of Metallurgy, The University of Tokyo, Hongo, Bunkyo, Tokyo 113-8656 Japan.

(Received on March 7, 2000; accepted in final form on June 13, 2000)

Thermodynamic properties of chlorine in the CaO–SiO2–Al2O3, Na2O–SiO2–Al2O3 and CaO–SiO2–Al2O3–Na2O slags have been investigated in the present study. The experiments were carried out using gas-slag equilibrium, controlling both \( P_{O_2} \) and \( P_{Cl_2} \) simultaneously (\( P_{O_2} = 10^{-16} - 10^{-13} \) atm, \( P_{Cl_2} = 10^{-10} - 10^{-12} \) atm at 1673–1748 K), and the solubility of chlorine in the 40mass%CaO–40mass%SiO2–20mass%Al2O3 slag was found to vary in proportion to \( P_{O_2}^{1/4} \) and \( P_{Cl_2}^{1/2} \). Accordingly, the chloride capacity (\( C_{Cl^-} \)), which represents the ability of slags to absorb chlorine, has been defined as:

\[
C_{Cl^-} = \frac{\text{mass\%Cl}^-}{P_{Cl_2}^{1/2}P_{O_2}^{1/4}}
\]

The \( C_{Cl^-} \) values were observed to increase with increasing slag basicity and temperature, and showed a reasonable relationship with that of \( C_{S^{2-}} \).

KEY WORDS: iron- and steelmaking slags; gas–slag equilibrium; chlorine; solubility of chlorine; chloride capacity; sulfide capacity; slag basicity.

1. Introduction

Ecological problems have recently been treated in iron- and steelmaking processes, as seen in the development of waste plastic injection into a blast furnace1). However, we have to develop more effective and more extensive utilization of such a high temperature mass production system for the waste management. At present, waste plastics containing chlorine such as polyvinyl chloride (PVC) cannot be treated due to the problem of dioxine and HCl generation as well as the erosion of refractories. In order to develop the waste treatment system for the materials containing chlorine, its behavior in high temperature furnaces must be predicted and the evaluation of chlorine gas absorption into slags becomes essential, which is also very important for practical incineration processes. However, no data are available regarding the thermodynamic properties of chlorine in molten slags, because it has not been treated as impurities in molten iron such as S, P and N.

In the present study, by measuring the chlorine concentrations at various compositions of the CaO–SiO2–Al2O3, Na2O–SiO2–Al2O3 and CaO–SiO2–Al2O3–Na2O slags in a controlled atmosphere (\( P_{O_2} \) and \( P_{Cl_2} \)), the dissolution mechanism of chlorine into molten slags has been clarified and the ability of slags to capture chlorine has been evaluated in terms of chloride capacity, \( C_{Cl^-} \).

2. Experimental

A SiC resistance furnace with a fused alumina tube (53 mm ID and 1000 mm length) was employed and experimental temperature (1673–1748 K) was controlled by a proportional-integral differential (PID) controller and kept within ±1 K with a Pt–6%Rh/Pt–30%Rh thermocouple. Schematic sketch inside the furnace is drawn in Fig. 1.

During experiments, the atmosphere was controlled by flowing a gas mixture of CO, H2, HCl and Ar at a rate of 50–150 Ncm3/min. As graphite crucibles were used, \( P_{O_2} \) was determined by Eqs. (1) and (2), and \( P_{Cl_2} \) by Eqs. (3) and (4), respectively.

![Fig. 1. Schematic cross section of the experimental furnace.](image-url)
Fig. 2. Schematic sketch of gas purification train.

\[ \Delta G^\circ = -RT \ln \frac{p^{1/2}}{P_{CO}} = 114\,600 + 85.9\,T \text{ [J/mol]} \]  \hspace{1cm} (2)

\[ \text{HCl(g)} = \frac{1}{2}\text{H}_2\text{(g)} + \frac{1}{2}\text{Cl}_2\text{(g)} \]  \hspace{1cm} (3)

\[ \Delta G^\circ = -RT \ln \frac{p^{1/2}}{p_{HCl}} = 94\,230 + 6.41T \text{ [J/mol]} \]  \hspace{1cm} (4)

In order to control partial pressures of gas species precisely, capillary mass flow units, as schematically shown in Fig. 2, were used. In the gas purification system, H$_2$SO$_4$ and Mg(ClO$_4$)$_2$ were used to remove H$_2$O, Mg turnings at 773 K for evaporation of Na$_2$O in the slags was inevitable due to the reaction with graphite crucibles, molten metals (Pb–Na) coexisted in order to supply its evaporation loss for several experiments. In order to confirm the equilibrium, some experiments were carried out for various periods, such as 6, 12, 18, 24 and 30 h. Consequently, the experimental time was determined to be 24 h for the CaO–SiO$_2$–Al$_2$O$_3$ and CaO–SiO$_2$–Al$_2$O$_3$–Na$_2$O slags, and 16 h for the Na$_2$O–SiO$_2$–Al$_2$O$_3$ slags. Just before taking out the samples, the experimental gas mixture was switched to Ar gas for 1–2 min. The samples were withdrawn and quenched in the flushing Ar gas and subjected to chemical analyses.

The CaO and Al$_2$O$_3$ contents of the slags were determined by inductively coupled plasma (ICP) emission spectrometry at wavelength of 393.4 nm and 396.2 nm, respectively. The Na$_2$O contents of the slags were determined by atomic absorption analysis at wavelength of 589 nm. The SiO$_2$ and total chlorine contents were determined by gravimetry and silver nitrate titration, respectively.

The slag compositions were corrected so that the total contents of the CaO, SiO$_2$, Al$_2$O$_3$ and Na$_2$O may become 100 mass%. Namely, the chlorine contents are excluded in representing the slag compositions after the experiments.

3. Results and Discussion

3.1. CaO–SiO$_2$–Al$_2$O$_3$ Slag System

Experimental results are summarized in Table 1.

3.1.1. Chlorine Dissolution into Molten Slag

In order to investigate the behavior of chlorine in molten slags under reducing conditions, the effects of $P_{Cl_2}$ and $P_{O_2}$ on the solubility of chlorine in the slag were investigated. The 40mass%CaO–40mass%SiO$_2$–20mass%Al$_2$O$_3$ slag was examined at $P_{Cl_2}$ and $P_{O_2}$ ranges from approximately 10$^{-13}$ to 10$^{-11}$ atm and 10$^{-19}$ to 10$^{-17}$ atm, respectively at 1673 K.

As seen in Fig. 3, there is a linear relationship between $\log P_{O_2}$ and $\log(mass\%Cl^-)/P_{Cl_2}^{1/2}$, and the slope of the line is found to be $-0.19$ by a linear regression, which is close to $-1/4$. Accordingly, the solubility of chlorine in the slag is considered to be proportional to $P_{O_2}^{-1/4}$ and $P_{Cl_2}^{1/2}$.

Based on this result, the dissolution mechanism of chlorine into the slag at the present experimental condition can be described by Eq. (5).

$$\frac{1}{2}\text{Cl}_2\text{(g)} + \frac{1}{2}\text{O}_2^-\text{(slag)} = \text{Cl}^-\text{(slag)} + \frac{1}{4}\text{O}_2\text{(g)} \quad (5)$$

This means that Cl$_2$ dissolves into the slag as Cl$^-$ ion and oxychloride ions may not be stable in such low oxygen partial pressures. Herewith, chloride capacity $C_{Cl^-}$ is defined from Eq. (5) as Eq. (6),

$$C_{Cl^-} = \frac{(mass\%Cl^-)P_{O_2}^{1/4}}{f_{Cl}} = K_{Cl}a^{1/2}_{Cl^-}$$  \hspace{1cm} (6)

where $K_{Cl}$ is the equilibrium constant of Eq. (5), $a_{Cl^-}$ is the activity of oxygen ion, and $f_{Cl}$ is the activity coefficient of chloride ion in the slag, respectively. Since the most right hand side of Eq. (6) is a constant at given temperature and slag composition, $C_{Cl^-}$ is a characteristic value of the slag composition.

3.1.2. Dependence of $C_{Cl^-}$ on Slag Composition

The obtained values for $C_{Cl^-}$ are plotted in the isothermal cross section of the CaO–SiO$_2$–Al$_2$O$_3$ system at 1748 K in Fig. 4. As seen in the figure, $C_{Cl^-}$ increases as the CaO content increases, and decreases as the SiO$_2$ and Al$_2$O$_3$ contents increase, which is consistently observed for both two liquid regions. The present result shows that $C_{Cl^-}$ increases as the slag basicity increases, and Al$_2$O$_3$ is considered to act as an acidic component in the present conditions. Therefore, the basicity index has been defined as Eq. (7),
where $\alpha$ is a parameter, which makes AlO$_{1.5}$ equally effective as SiO$_2$ for lowering the chloride capacity. It was found, from calculations using the experimental data obtained, to be 0.67.

Figure 5 shows the relationship between $C_{\text{Cl}^-}$ and $B$. As is seen in the figure, the $C_{\text{Cl}^-}$ value can be roughly expressed by a linear equation of $B$ as shown in Eq. (8).

$$C_{\text{Cl}^-} = -1.8 + 42.5B$$

where $B = \frac{X_{\text{CaO}}}{X_{\text{SiO}_2} + \alpha X_{\text{AlO}_2}}$; $\alpha = 0.67$ ........................ (7)

3.1.3. Relationship between $C_{\text{Cl}^-}$ and $C_{\text{S}^{2-}}$

Sulfide capacity ($C_{\text{S}^{2-}}$) is also known as the capability of slags to absorb sulfur based on the reaction shown in Eq. (9), and it is defined by Eq. (10).\(^3\)

$$\frac{1}{2} S_2(g) + O^2-(\text{slag}) = S^{2-}(\text{slag}) + \frac{1}{2} O_2(g) \text{ .......... (9)}$$

$$C_{\text{S}^{2-}} = \frac{\text{mass}\% S^{2-}}{P_{O_2}^{1/2}} \frac{P_{O_2}^{1/2}}{P_{S_2}^{1/2}} = K_s \frac{O^2-}{f_{O^2-}} \text{ .......... (10)}$$

The $C_{\text{S}^{2-}}$ values for the CaO–SiO$_2$–Al$_2$O$_3$ slags have been...
measured by a number of researchers. Among them, the \( C_{S^2} \) values at 1773 K, which is close to the experimental temperature of 1748 K, were measured by Kalyanram et al. As shown in Fig. 6. Although \( C_{S^2} \) as well as \( C_{Cl^2} \) increases as the slag basicity increases, \( C_{S^2} \) seems to be more sensitive to slag composition when compared with \( C_{Cl^2} \) in Fig. 4.

Substituting Eq. (6) into Eq. (10), one can derive the relationship between \( C_{Cl^2} \) and \( C_{S^2} \) as Eq. (11).

\[
\log C_{Cl^2} = \frac{1}{2} \log C_{S^2} + \log \frac{K_1}{K_3^{1/2}} + \log \frac{f_{S^2}^{1/2}}{f_{Cl^2}} \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ ld
respectively. Using these values, the contribution of dissolution of diatomic molecules in the Eqs. (5) and (8) are calculated to be $2.1 \text{kJ/mol}$ and $37 \text{kJ/mol}$, respectively. Considering that the difference of these two values is small enough compared to the $D_{\text{H}}$ values of Eqs. (5) and (8), the mechanism of dissolution of chlorine into slags may be similar to that of sulfur.

3.2. Na$_2$O–SiO$_2$–Al$_2$O$_3$ Slag System

Generally speaking, soda based slags have higher basicity than lime based slags. It has been also reported that sulfide capacities for the 3Na$_2$O–2SiO$_2$–3CaO–2SiO$_2$ and the Na$_2$O–SiO$_2$–CaO–SiO$_2$ pseudo-binary systems decreases with increasing 3CaO–2SiO$_2$ and CaO–SiO$_2$ contents. In the present study, the $C_{\text{Cl}}$ values for the Na$_2$O–SiO$_2$–Al$_2$O$_3$ slags are also measured at 1673 K (Table 3). The obtained values are plotted in the isothermal cross section of the Na$_2$O–SiO$_2$–Al$_2$O$_3$ system at 1673 K in Fig. 9. As seen in the figure, the $C_{\text{Cl}}$ values for the Na$_2$O–SiO$_2$–Al$_2$O$_3$ slags are significantly different from those for the CaO–SiO$_2$–Al$_2$O$_3$ slags because Na$_2$O is more basic oxide than CaO.

3.3. CaO–SiO$_2$–Al$_2$O$_3$–Na$_2$O Slag System

The effect of the Na$_2$O addition on $C_{\text{Cl}}$ for the CaO–SiO$_2$–Al$_2$O$_3$ slags was investigated. (Table 4) All the experiments were carried out with Pb–1mass%Na alloy in the crucible. Two compositions were chosen for base slags, 40mass%CaO–40mass%SiO$_2$–20mass%Al$_2$O$_3$ and 20mass%CaO–65mass%SiO$_2$–15mass%Al$_2$O$_3$, respectively, and various amount of Na$_2$O was added to the slags. The results are shown in Fig. 11 as a function of Na$_2$O content of the final slags. The $C_{\text{Cl}}$ values significantly increased by Na$_2$O addition because of the increase in the slag basicity, which is similar to the effect of Na$_2$O addition to the CaO–SiO$_2$–CaF$_2$ slags on its phosphate capacity by Muraki et al. Moreover, it is considered larger increase in $C_{\text{Cl}}$ was observed for the higher CaO slags.

In the present study, the $C_{\text{Cl}}$ values obtained at 1673 K were expressed as an empirical approximation of the contents of components. In order to make it available for both ternary and quaternary systems, the multiplication term of basic components ($X_{\text{CaO}}X_{\text{NaO}}^{0.5}$) was introduced. The basicity index $B$ has been modified as Eq. (12), assuming the equ-
fect of Al$_2$O$_3$ as an acidic oxide at 1673 K is the same as that of 1748 K.

\[
B = \frac{X_{\text{CaO}} + \beta X_{\text{NaO}} + \gamma X_{\text{CaO}} X_{\text{NaO}}}{X_{\text{SiO}} + 0.67 X_{\text{AlO}}}; \quad \beta = 6, \quad \gamma = 27
\]

where parameters $\beta$ and $\gamma$ were determined in terms of trial and error method. Figure 12 shows the relationship between $C_{\text{Cl}}$ and B. All the plots in the figure fit the broken line with a slope of 10 with an intersection at 7.2 with vertical axis. Therefore, the $C_{\text{Cl}}$ values is approximated by the following empirical equation;

\[
C_{\text{Cl}} = 7.2 + 10B
\]

\[
= 7.2 + \frac{10X_{\text{CaO}} + 60X_{\text{NaO}} + 270X_{\text{CaO}} X_{\text{NaO}}}{X_{\text{SiO}} + 0.67 X_{\text{AlO}}} \quad \text{(1673K)}
\]

\[
.........................................(13)
\]

4. Conclusions

The solubility of chlorine in the molten CaO–SiO$_2$–Al$_2$O$_3$, Na$_2$O–SiO$_2$–Al$_2$O$_3$ and CaO–SiO$_2$–Al$_2$O$_3$–Na$_2$O slags have been measured under reducing conditions by a gas-slag equilibration technique, yielding the following results:

1) The solubility of chlorine in the slag is proportional to both $P_{\text{Cl}}^{1/4}$ and $P_{\text{Cl}}^{1/2}$, showing that chlorine dissolves into the slag as Cl$^-$ ion, and $C_{\text{Cl}}$ is defined as;

\[
C_{\text{Cl}} = \frac{\text{mass}\% \text{Cl}^-}{P_{\text{Cl}}^{1/4}}
\]

2) For the CaO–SiO$_2$–Al$_2$O$_3$ slags, the $C_{\text{Cl}}$ value increases as the CaO content increases, and decreases as the SiO$_2$ and Al$_2$O$_3$ contents increase, which is shown by a linear relationship between $C_{\text{Cl}}$ and the basicity index $B$, $X_{\text{CaO}}/(X_{\text{SiO}} + 0.67X_{\text{AlO}})$.  

3) The $C_{\text{Cl}}$ value is less affected by the composition of slags than that of $C_{\text{Cl}}$. In the liquid region with higher SiO$_2$ content, there is a good linear correlation between $\log C_{\text{Cl}}$ and $\log C_{\text{Cl}}$ with a slope of approximately 1/2, which is consistent with the value predicted from their definitions on the assumption of the constancy of the value for $f_{\text{Cl}}^{12}/f_{\text{Cl}}^{1/2}$.

4) The $C_{\text{Cl}}$ value increases as the temperature increases. The $\Delta H^\circ$ value of chlorine dissolution into each slag composition calculated from the slopes of the temperature dependence increases as B increases.

5) For the Na$_2$O–SiO$_2$–Al$_2$O$_3$ slags, the $C_{\text{Cl}}$ value increases as the Na$_2$O content increases. The $C_{\text{Cl}}$ values for the Na$_2$O–SiO$_2$–Al$_2$O$_3$ slags are higher than those for the CaO–SiO$_2$–Al$_2$O$_3$ slags because Na$_2$O is more basic oxide than CaO.

6) The $C_{\text{Cl}}$ value for the CaO–SiO$_2$–Al$_2$O$_3$ slags increases by Na$_2$O addition. The larger increase in $C_{\text{Cl}}$ was observed for the higher CaO slags.

7) The $C_{\text{Cl}}$ values for the three kinds of slags at 1673 K were approximated by the following empirical equation:

\[
C_{\text{Cl}} = 7.2 + \frac{10X_{\text{CaO}} + 60X_{\text{NaO}} + 270X_{\text{CaO}} X_{\text{NaO}}}{X_{\text{SiO}} + 0.67 X_{\text{AlO}}}
\]

Acknowledgements

This work was performed in the framework of “Research for the Future” Program of the Japan Society for the Promotion of Science. (JSPS-RFTF96R01901)

REFERENCES

1) K. Wakimoto, H. Nakamura, M. Fujii, Y. Yamada, K. Nemoto and K. Tomioka: Nippon Kokan Tech. Rep., 160 (1997), 1.
2) E.T.Turkdogan: Physical Chemistry of High Temperature Technology, Academic Press, New York, (1980), 5.
3) F. D. Richardson and C. J. B. Fincham: J. Iron Steel Inst., 178 (1954), 4.
4) R. A. Sharma and F. D. Richardson: J. Iron Steel Inst., 198 (1961), 386.
5) R. A. Sharma and F. D. Richardson: J. Iron Steel Inst., 200 (1962), 373.
6) M. R. Kalyanram, T. G. MacFarlane and H. B. Bell: J. Iron Steel Inst., 195 (1960), 58.
7) M. Görnerup and O. Wijk: Scand. J. Metall., 25 (1996), 103.
8) M. W. Chase, Jr.: NIST-JANAF Thermochemical Tables Fourth Edition, (1998), 737, 1717 and 1864.
9) R. Inoue and H. Suito: Trans. Iron Steel Inst. Jpn., 22 (1982), 515.
10) M. Muraki, H. Fujikuma and N. Sano: Trans. Iron Steel Inst. Jpn., 25 (1985), 1025.