

A THERMODYNAMIC STUDY ON THE OXIDATION OF SILICON, CARBON AND CHROMIUM IN THE FERROCHROME CONVERTER

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ABSTRACT

The purpose of a ferro-chrome converter (hereafter abbreviated as CRK) is to remove silicon as well as part of carbon from the molten ferro-chrome before the production of stainless steel by blowing oxygen into the melt. The product of the CRK is a raw material for the AOD converter and stainless steel production, whereas the raw materials of the CRK are molten ferro-chrome with high silicon and carbon contents (from the ferro-chrome plant) and stainless steel scrap that is used to decrease the melt's temperature that is increased due to oxidation reactions of silicon and carbon. The CRK process could therefore be considered as a link between the ferro-chrome production and the production of stainless steels.

The oxidation reactions of silicon, carbon and chromium in the CRK process were considered in this study by comparing the results of computational thermodynamics with the slag and steel analyses that were taken from the Outokumpu Stainless Tornio steelworks. The results indicate that the oxidation reactions of silicon, carbon and chromium in the end of the blowing are not in mutual equilibrium.

1 INTRODUCTION

Computational thermodynamics has proved to be a suitable tool in the control, modelling and understanding of pyrometallurgical processes. On the other hand, the process data acquired from the measurements from the metallurgical processes has been widely used in the validation of the thermodynamic models and thermochemical data. There is, however, less information available concerning the CRK process and its thermodynamic state in comparison with some other metallurgical unit operations, since it is not as widely used as e.g. blast furnace or basic oxygen furnace.

The purpose of this study was to compare computational values for thermodynamic equilibria with process data measurements in order to estimate whether the CRK process is in equilibrium or not in the final stage of its blowing. Since carbon, silicon and chromium are the most essential elements in the CRK melts (in addition to iron), the reactions between these elements were chosen as a main topic of this study.

2 FERROCHROME CONVERTER

The task of the CRK process is to serve as a liquid ferrochromium buffer between ferrochrome plant and stainless steel melting shop; as well as to utilise chemical energy of ferrochromium for scrap melting. With the CRK process the use of liquid FeCr in the melting shop process is maximised and it also decreases the need for electric energy in EAF. In addition to this, it is possible to replace an AOD converter with CRK in the possible case of a AOD breakdown.

Liquid ferrochrome contains approximately 4 w-% silicon and 7 w-% carbon. All silicon and some of carbon are oxidized in CRK process by blowing pure oxygen or oxygen/compressed air -mixture into the bath. Thermal energy that is generated in the oxidation reactions is utilized for scrap and alloy

melting. During the process, 550 kg of scrap and 150 kg of slag formers per ton of produced ferrochromium are melted whereas the charge temperature is risen up to 1650 - 1700 °C. Typical initial and final compositions of the CRK charge are shown in Table 1 and the analysis of slag after CRK process is presented in Table 2. [1]

Table 1: Melt composition and temperature at the beginning and at the end of CRK process [1].

	Start	Final
Cr (w-%)	53	35
Ni (w-%)	0.2	5
C (w-%)	7	3
Si (w-%)	4	0.3
T (°C)	1530	1650-1700

Table 2: Typical analysis of CRK slag after processing [1].

CaO	SiO ₂	MgO	FeO	Cr ₂ O ₃	Al ₂ O ₃	TiO ₂	MnO	CaO/SiO ₂
(w-%)	(w-%)	(w-%)	(w-%)	(w-%)	(w-%)	(w-%)	(w-%)	
42-45	34-38	9-12	1-2	2-3	2-3	1-2	0.5-1	1.2-1.4

The oxidation of silicon, carbon and chromium is illustrated in Figure 1 in which the Cr₂O₃-content of the slag as well as the Si- and C-contents of the metal are shown as a function of the amount of blown oxygen [1].

Nominal capacity of the CRK process in Outokumpu Stainless Tornio Works is 95 tons and the geometry of the vessel is similar to AOD converter at the same plant and it is lined with MgO-C bricks. Normally the charge weight varies from 30 to 100 tons. The CRK process is equipped with 5 side-mounted tuyeres located at 40 centimeters height from the vessel bottom. Top lance is Laval type with three holes (3HL). In the beginning of the blowing approximately 60 m³/min (STP) of oxygen is blown through the tuyeres (cf. stage 6 in Figure 1). In stage 22 (cf. Figure 1) approximately 50 m³/min (STP) of air is blown through the tuyeres in addition to approximately 130 m³/min (STP) of oxygen that is blown through the lance. The final stage is similar to the first stage with the oxygen blowing of approximately 60 m³/min (STP) through the tuyeres. [1]

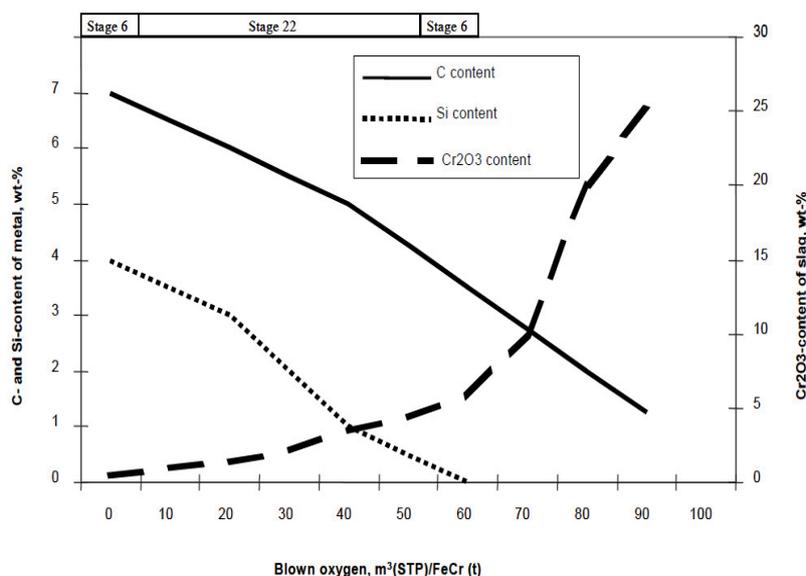


Figure 1: Changes of slag and metal phase compositions during blowing in CRK process [2].

After the blowing a metal sample is taken through slag with lollipop type sampling probe and slag sample with iron rod. Both samples are taken manually from tilted CRK. Carbon contents of the metal sample are analyzed with optical emission spectrometer (ARL-4460) and other elements with XRF

(Panalytical Axios Advanced). Grinded slag briquettes are analyzed with the same XRF as steel samples.

3 SLAG-METAL EQUILIBRIUM

3.1 Equations

In order to define correlations between the amounts of chromium, silicon and carbon in molten metal and slag, the reactions presented in Equations (1) to (3) were chosen to be considered in the computations. The equilibrium constants for the reactions presented in Equations (1) to (3) are presented in Equations (4) to (6), respectively.



$$K_{(1)} = \frac{a_{SiO_2}^3 \cdot a_{Cr}^4}{a_{Si}^3 \cdot a_{Cr_2O_3}^2} = \frac{a_{SiO_2}^3 \cdot f_{Cr}^4 \cdot X_{Cr}^4}{f_{Si}^3 \cdot X_{Si}^3 \cdot a_{Cr_2O_3}^2} = e^{-\left(\frac{\Delta G_{(1)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \quad (4)$$

$$K_{(2)} = \frac{p_{CO}^2 \cdot a_{Si}}{a_C^2 \cdot a_{SiO_2}} = \frac{p_{CO}^2 \cdot f_{Si} \cdot X_{Si}}{f_C^2 \cdot X_C^2 \cdot a_{SiO_2}} = e^{-\left(\frac{\Delta G_{(2)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \quad (5)$$

$$K_{(3)} = \frac{p_{CO}^3 \cdot a_{Cr}^2}{a_C^3 \cdot a_{Cr_2O_3}} = \frac{p_{CO}^3 \cdot f_{Cr}^2 \cdot X_{Cr}^2}{f_C^3 \cdot X_C^3 \cdot a_{Cr_2O_3}} = e^{-\left(\frac{\Delta G_{(3)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \quad (6)$$

for which a_i represents the activity of component i , p_i the partial pressure of component i , X_i the mole fraction of component i and f_i the activity coefficient of component i . $\Delta G_{(n)}^0$ equals the standard Gibbs energy for the reaction presented in Equation (n), $\Delta G_{R \rightarrow H}$ equals the Gibbs energy for the change between the Raoultian and Henrian standard states, R equals the gas constant and T equals temperature.

3.2 Computational methods and assumptions

The relations between $a_{Cr_2O_3}$ and X_{Si} , a_{SiO_2} and X_C as well as $a_{Cr_2O_3}$ and X_C were calculated using Equations (7) to (9), that were obtained from the Equations (4) to (6), respectively.

$$a_{Cr_2O_3} = \sqrt{\frac{a_{SiO_2}^3 \cdot f_{Cr}^4 \cdot X_{Cr}^4}{f_{Si}^3 \cdot X_{Si}^3} \cdot e^{-\frac{\Delta G_{(1)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}}} \quad (7)$$

$$a_{SiO_2} = \frac{p_{CO}^2 \cdot f_{Si} \cdot X_{Si}}{f_C^2 \cdot X_C^2} \cdot e^{-\frac{\Delta G_{(2)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}} \quad (8)$$

$$a_{Cr_2O_3} = \frac{p_{CO}^3 \cdot f_{Cr}^2 \cdot X_{Cr}^2}{f_C^3 \cdot X_C^3} \cdot e^{-\frac{\Delta G_{(3)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}} \quad (9)$$

In order to be able to calculate the activities of Cr_2O_3 and SiO_2 as a function of X_{Si} and X_C , the values for other parameters presented in Equations (7) to (9) needed to be assumed or calculated using other methods. The values used in the calculations within this study as well as the list of used models and assumptions are presented in Table 3.

Table 3: The values used in the calculations as well as the list of used models and assumptions.

Parameter in Equation			Used value(s)	Models and assumptions that were used
(7)	(8)	(9)		
a_{SiO_2}			0.11	An average value that was achieved by calculating the activities of SiO_2 with the quadratic formalism by Ban-Ya [3] and Xiao <i>et al.</i> [4] and using measured SiO_2 -contents of the CRK-slag at the end of blowing as starting values.
f_{Cr}			0.637-0.643 *	Calculated as a function of X_{Si} by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state).
X_{Cr}		X_{Cr}	0.347	An average value of the measured chromium contents of the metal at the end of blowing.
f_{Si}			3.31-3.69 *	Calculated as a function of X_{Si} by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state).
$\Delta G^0_{(1)}$			21.16·T - 134700 (cal/mol)	Standard free energy change for the reaction presented in Equation (1) achieved from the database of the HSC Chemistry for Windows, V6.1 [6].
$\Delta G_{\text{R} \rightarrow \text{H}}$			66200 (cal/mol)	4·R·T·ln f^0_{Cr} - 3·R·T·ln f^0_{Si} (values of f^0_{Cr} and f^0_{Si} achieved from reference [7]).
	p_{CO}	p_{CO}	1.9; 2.15; 2.4 (bar)	Estimated ferrostatic pressures in three different depths of the melt in the CRK process.
	f_{Si}		1.87-4.56 **	Calculated as a function of X_{C} by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state).
	X_{Si}		0.002	An average value of the measured silicon contents of the metal at the end of blowing.
	f_{C}		0.369-1.41 **	Calculated as a function of X_{C} by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state).
	$\Delta G^0_{(2)}$		-87.40·T +145900 (cal/mol)	Standard free energy change for the reaction presented in Equation (2) achieved from the database of the HSC Chemistry for Windows, V6.1 [6].
	$\Delta G_{\text{R} \rightarrow \text{H}}$		-17900 (cal/mol)	R·T·ln f^0_{Si} - 2·R·T·ln f^0_{C} (values of f^0_{Si} and f^0_{C} achieved from reference [7]).
		f_{Cr}	0.545-0.993 **	Calculated as a function of X_{C} by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state).
		f_{C}	0.166-1.411 **	Calculated as a function of X_{C} by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state).
		$\Delta G^0_{(3)}$	-120.5·T +151400 (cal/mol)	Standard free energy change for the reaction presented in Equation (3) achieved from the database of the HSC Chemistry for Windows, V6.1 [6].
		$\Delta G_{\text{R} \rightarrow \text{H}}$	6300 (cal/mol)	2·R·T·ln f^0_{Cr} - 3·R·T·ln f^0_{C} (values of f^0_{Cr} and f^0_{C} achieved from reference [7]).
* depends on the value of X_{Si}				
** depends on the value of X_{C}				

3.3 Results

The calculated values of activities of Cr_2O_3 and SiO_2 as a function of X_{Si} and X_{C} are presented in Figures 2 to 4 together with the measured melt compositions and calculated slag activity values from the CRK process of the Outokumpu Stainless Tornio works. Activities of Cr_2O_3 and SiO_2 from slag samples were calculated with the quadratic formalism by Ban-Ya [3] and Xiao *et al.* [4].

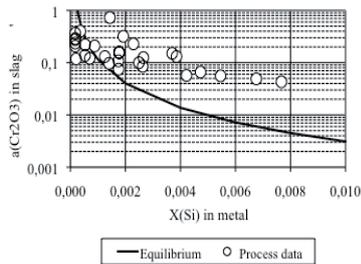


Figure 2: Calculated activity of Cr_2O_3 in the slag as a function of silicon content in the metal as well as measured process data from the CRK process.

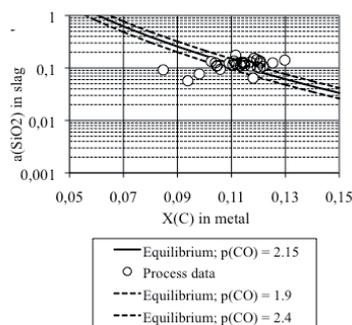


Figure 3: Calculated activity of SiO_2 in the slag as a function of carbon content in the metal as well as measured process data from the CRK process.

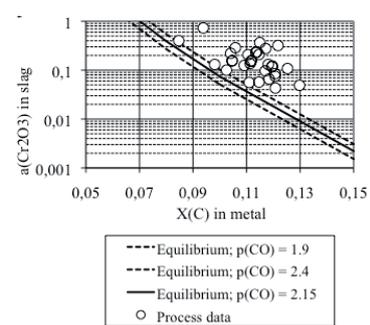


Figure 4: Calculated activity of Cr_2O_3 in the slag as a function of carbon content in the metal as well as measured process data from the CRK process.

3.4 Discussion

It is seen from Figure 2 that the Cr_2O_3 -activity in the slag depends on the metal's silicon content according to the equilibrium of the reaction (1). Activity of Cr_2O_3 in slag increases as the silicon content in melt is lowered (cf. Figure 1). When the silicon content of the metal is close to zero (as it usually is at the end of the blowing), the calculated activity of Cr_2O_3 closes to unity. Calculated equilibrium and the measured process data are in good agreement in low but not at high silicon contents. This indicates that slag in CRK process has not been in thermodynamic equilibrium at the end of the blowing. Since high silicon contents in the metal are not desirable at the end of the CRK-blowing, it is likely that concerning these measurements there has been some sort of deviation from the normal practice due to which the equilibrium has not been reached. The most probable explanation is insufficient mixing concerning these measurements. In order to get a more accurate image concerning the attainment of equilibrium in the CRK process, a comparison should be made between the thermodynamic calculations and process measurements taken from start, middle and end of the CRK process.

Small deviations in the activity of SiO_2 as a function of the metal's carbon content are seen from Figure 3. The relatively constant values of a_{SiO_2} can be explained with the use of suitable amounts of CaO and MgO as slag modifiers in order to keep the slag basicity in control. If SiO_2 activity in slag would be higher, e.g. with slag composition modification, lower carbon contents in melt could be achieved. On other hand carbon content in melt has a direct effect to chromium oxidation in equilibrium as can be seen from Figure 4.

4 PARTIAL PRESSURE OF OXYGEN

4.1 Reactions

In order to estimate whether the oxidation reactions of silicon, carbon and chromium are in mutual equilibrium, these reactions (presented in equations (10) to (12)) were considered also separately. The equilibrium constants for reactions presented in equations (10) to (12) are presented in equations (13) to (15), respectively.





$$K_{(10)} = \frac{a_{SiO_2}}{a_{Si} \cdot p_{O_2}} = \frac{a_{SiO_2}}{f_{Si} \cdot X_{Si} \cdot p_{O_2}} = e^{-\left(\frac{\Delta G_{(10)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \quad (13)$$

$$K_{(11)} = \frac{a_{Cr_2O_3}}{a_{Cr}^2 \cdot p_{O_2}^{3/2}} = \frac{a_{Cr_2O_3}}{f_{Cr}^2 \cdot X_{Cr}^2 \cdot p_{O_2}^{3/2}} = e^{-\left(\frac{\Delta G_{(11)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \quad (14)$$

$$K_{(12)} = \frac{p_{CO}^2}{a_C^2 \cdot p_{O_2}} = \frac{p_{CO}^2}{f_C^2 \cdot X_C^2 \cdot p_{O_2}} = e^{-\left(\frac{\Delta G_{(12)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \quad (15)$$

for which a_i represents the activity of component i , p_i the partial pressure of component i , X_i the mole fraction of component i and f_i the activity coefficient of component i . $\Delta G_{(n)}^0$ equals the standard Gibbs energy for the reaction presented in Equation (n), $\Delta G_{R \rightarrow H}$ equals the Gibbs energy for the change between the Raoultian and Henrian standard states, R equals the gas constant and T equals temperature.

4.2 Computational methods and assumptions

Partial pressures of oxygen corresponding the equilibrium between silicon and SiO_2 , chromium and Cr_2O_3 as well as carbon and CO were calculated using Equations (16) to (18), that were obtained from the Equations (13) to (15), respectively.

$$p_{O_2} = \frac{a_{SiO_2}}{f_{Si} \cdot X_{Si}} \cdot e^{-\left(\frac{\Delta G_{(10)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \quad (16)$$

$$p_{O_2} = \left[\frac{a_{Cr_2O_3}}{f_{Cr}^2 \cdot X_{Cr}^2} \cdot e^{-\left(\frac{\Delta G_{(11)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \right]^{2/3} \quad (17)$$

$$p_{O_2} = \frac{p_{CO}^2}{f_C^2 \cdot X_C^2} \cdot e^{-\left(\frac{\Delta G_{(12)}^0 + \Delta G_{R \rightarrow H}}{R \cdot T}\right)} \quad (18)$$

The values needed for the calculations of the partial pressures as well as the assumptions behind the values are presented in Table 4.

Table 4: The values used in the calculations as well as the list of used models and assumptions.

Parameter in Equation			Used value(s)	Models and assumptions that were used
(16)	(17)	(18)		
a_{SiO_2}			0.08-0.18	Calculated SiO_2 activities with the quadratic formalism by Ban-Ya [3] and Xiao <i>et al.</i> [4] from measured contents of the CRK-slag at the end of blowing.
f_{Si}			2.52-4.20	Calculated by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state) for different metal compositions.
X_{Si}			0.00018-0.0077	Measured Si-contents of the metal at the end of blowing.
$\Delta G^0_{(10)}$			47.2·T - 213000 (cal/mol)	Standard free energy change for the reaction presented in Equation (10) achieved from the database of the HSC Chemistry for Windows, V6.1 [6].
$\Delta G_{\text{R} \rightarrow \text{H}}$			25100 (cal/mol)	$-R \cdot T \cdot \ln f^0_{\text{Si}}$ (value of f^0_{Si} achieved from reference [7]).
	$a_{\text{Cr}_2\text{O}_3}$		0.05-0.73	Calculated Cr_2O_3 activities with the quadratic formalism by Ban-Ya [3] and Xiao <i>et al.</i> [4] from measured contents of the CRK-slag at the end of blowing.
	f_{Cr}		0.59-0.73	Calculated by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state) for different metal compositions.
	X_{Cr}		0.33-0.36	Measured Cr-contents of the metal at the end of blowing.
	$\Delta G^0_{(11)}$		60.2·T - 252000 (cal/mol)	Standard free energy change for the reaction presented in Equation (11) achieved from the database of the HSC Chemistry for Windows, V6.1 [6].
	$\Delta G_{\text{R} \rightarrow \text{H}}$		-990 (cal/mol)	$-2 \cdot R \cdot T \cdot \ln f^0_{\text{Cr}}$ (value of f^0_{Cr} achieved from reference [7]).
		p_{CO}	2.15 (bar)	Estimated ferrostatic pressure in the CRK process.
		f_{C}	0.59-1.20	Calculated by using the unified interaction parameter (UIP) formalism by Pelton and Bale [5] (Henrian standard state) for different metal compositions.
		X_{C}	0.085-0.13	Measured C-contents of the metal at the end of blowing.
		$\Delta G^0_{(12)}$	-40.2·T - 67400 (cal/mol)	Standard free energy change for the reaction presented in Equation (12) achieved from the database of the HSC Chemistry for Windows, V6.1 [6].
		$\Delta G_{\text{R} \rightarrow \text{H}}$	4250 (cal/mol)	$-2 \cdot R \cdot T \cdot \ln f^0_{\text{C}}$ (value of f^0_{C} achieved from reference [7]).

4.3 Results

Calculated partial pressures of oxygen corresponding the equilibrium between measured Si- and SiO_2 -contents, measured Cr- and Cr_2O_3 -contents as well as measured C-content and estimated partial pressure of CO are presented in Figures 5 to 7, respectively. Activities of Cr_2O_3 and SiO_2 from slag samples were calculated with the quadratic formalism by Ban-Ya [3] and Xiao *et al.* [4].

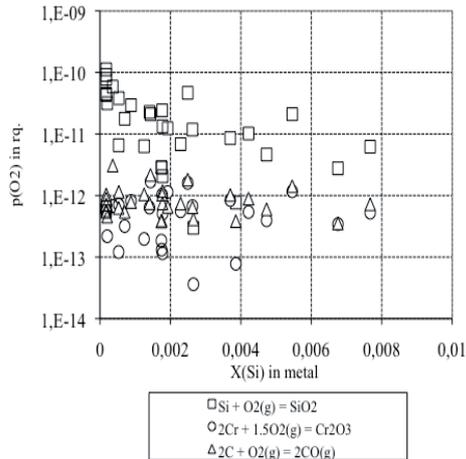


Figure 5: Partial pressures of oxygen as a function of silicon content in the metal calculated from the measured process data concerning the slag and metal compositions at the end of the blowing.

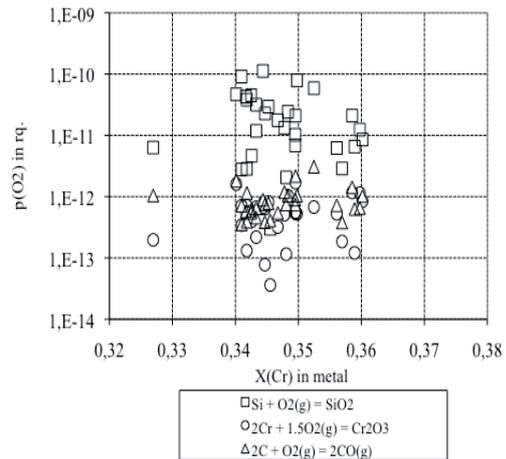


Figure 6: Partial pressures of oxygen as a function of chromium content in the metal calculated from the measured process data concerning the slag and metal compositions at the end of the blowing.

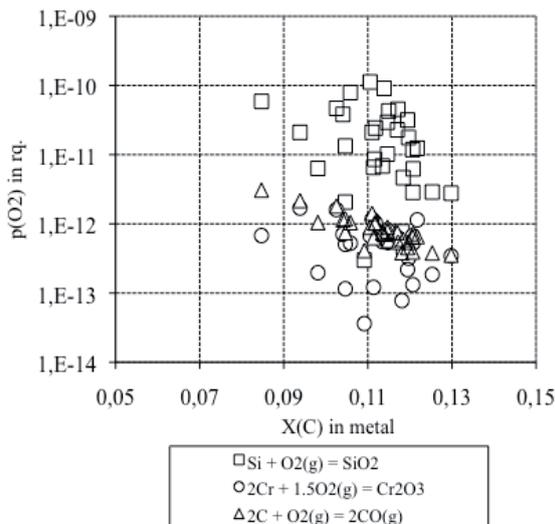


Figure 7: Partial pressures of oxygen as a function of carbon content in the metal calculated from the measured process data concerning the slag and metal compositions at the end of the blowing.

4.4 Discussion

The calculated partial pressures of oxygen presented in Figures 5 to 7 represent the order of oxidation for silicon, carbon and chromium at the end of the blowing (i.e. when the samples were taken from the CRK process). It is seen from these figures that since the silicon content of the metal approaches zero at the end of the blowing, the driving force for the oxidation of carbon and chromium becomes greater than the one for the oxidation of silicon (i.e. the partial pressures of oxygen

describing the equilibrium between carbon and CO as well as chromium and Cr_2O_3 are lower than the ones describing the equilibrium between silicon and SiO_2 in Figures 5 to 7), although the silicon is more easily oxidized at the beginning of the blowing (cf. Figure 1).

In Figure 5 it is shown that the partial pressure of oxygen in equilibrium with Si and SiO_2 depends clearly on the metal's silicon content, whereas Si-content's influence on the equilibrium between Cr and Cr_2O_3 as well as carbon and CO is insignificant. This makes it possible to estimate the silicon content below which it becomes more favourable for chromium and carbon to be oxidized in comparison with silicon. Based on Figure 5 this value is approximately 0,005 X(Si) or 0.3 w-%, whereas observations from the CRK process indicate that the oxidation of chromium is insignificant if the silicon content of the metal is above 0.1 w-%. In order to prevent chromium's oxidation below this silicon level, it is necessary to make adjustments to the blowing practices (i.e. partial blowing of nitrogen or argon instead of pure oxygen). The lack of mutual equilibrium between the oxidation reactions presented in equations (10) to (12) is also seen from Figures 6 and 7.

5 CONCLUSIONS

The oxidation of silicon, carbon and chromium in the CRK process was considered in this study by comparing computational results concerning the chemical equilibrium with the data that was achieved from the process measurements. Based on these comparisons it was concluded that the oxidation reactions are not in a mutual equilibrium at the end of the blowing and one should therefore be careful when verifying thermodynamic modelling with the process measurements. There could be several possible explanations to this finding from mass transfer controlled process to activity coefficients or reactions used.

In this paper, only trivalent state of chromium was considered as a slag product. Further work will be done to calculate equilibrium with divalent chromium oxide and to compare those results to process samples taken from start, middle and end of the CRK process.

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