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## Physical and chemical bases of decarburization of high-carbon ferromanganese melt

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### Фізичні та хімічні основи зневуглецювання високовуглецевого феромарганцевого розплаву

**Метою** роботи є встановлення фізико-хімічних закономірностей поведінки вуглецю, кремнію, марганцю при застосуванні методу продувки киснем високовуглецевого феромарганцю. **Методика.** Розглянуто процес продувки рідкого металу киснем. При продувці розплаву киснем відбувається переважне окислення кремнію. Його присутність в металі практично блокує окислення марганцю. Оскільки кисень є асиміляційним газом, процеси змішування компонентів ванни конвертера і відновлення оксидів марганцю на кордоні розділу метал-шлак не отримують належного розвитку під час продування.

Плавки середньовуглецевого феромарганцю в конвертері з дуттям характеризуються стабільним хімічним складом і майже повністю відповідають вимогам для цього типу сплаву. Невелике перевищення концентрації кремнію в металі по ряду плавок можна легко усунути шляхом додаткової продувки ванни киснем на заключній стадії рафінування. Поведінка фосфору в цих плавках не контролюється. Вміст  $P_2O_5$  в кінцевому шлаку становить 0,1%. Для досягнення прийнятних концентрацій фосфору в металі необхідно використовувати вихідні матеріали з низьким вмістом фосфору. **Наукова новизна.** Беручи до уваги високу спорідненість кремнію до кисню, фізичну та хімічну основу для виробництва середньовуглецевого феромарганцю, а також металевого марганцю та низьковуглецевого феромарганцю, відбувається процес взаємодії оксидів марганцю певний основний розплав шлаку з кремнієм, розчиненим у феромарганці (марганці), тобто як комбінований процес відновлення-рафінування для отримання феросплавів марганцю із заданим стандартом вмісту кремнію

**Ключові слова:** феромарганець, продувка киснем, фосфор, шлак, термодинаміка

**The aim** of the work is to establish physicochemical patterns of behavior of carbon, silicon, manganese when using the method of oxygen purge of high-carbon ferromanganese. **Method.** The process of blowing red metal to sour is neglected. With the fusion of fused acid, it is more important to oxidize silicon. Its presence in metal is practical in the block of oxidized manganese. Because oxygen is an assimilation gas, the mixing processes of the converter bath components and the reduction of manganese oxides at the metal-slag interface do not develop properly during purging.

The smelters of the medium-carbonaceous ferromanganese in the converter are characterized by a stable chemical warehouse and even a higher number of vimogs for this type of alloy. The low concentration of silicon in metal over a number of swimming trunks can be easily shoved with a hat of pre-purge bathtub with sour at the final stage of refining.

The behavior of phosphorus in these smelts is not controlled. The content of  $P_2O_5$  in the final slag is 0.1%. To achieve acceptable concentrations of phosphorus in the metal, it is necessary to use starting materials with a low phosphorus content. **Scientific novelty.** Taking into consideration the high affinity of silicon for oxygen, the physical and chemical basis for the production of medium-carbon ferromanganese, as well as metallic manganese and low-carbon ferromanganese, is the process of the interaction of manganese oxides of a certain basicity slag melt with silicon dissolved in ferromanganese (manganese), that is, as combined reduction -refining process to produce manganese ferroalloys with a given silicon content standard

**Key words:** ferromanganese, oxygen purge, phosphorus, slag, thermodynamics

#### Introduction.

Manganese ferroalloys, which are produced in Ukraine, are widely used in the smelting of steel and alloys, mainly in the form of ferromanganese and ferrosilicon manganese. When smelting a number of steel grades, it is necessary to use ferromanganese with a reduced carbon content - medium- and low-carbon ferromanganese.

The initial high-carbon ferromanganese for oxidative refining from carbon to obtain medium-carbon ferromanganese must have a low content of phosphorus and silicon. These requirements may be met by high-carbon ferromanganese produced with the use of low-phosphorous manganese ore charge and preliminarily refined from silicon with oxygen before pouring it into a converter for further decarburization. As previous studies have shown, medium-carbon ferromanganese after oxidative decarburization of high-carbon ferromanganese of the

common composition from Nikopol oxidic concentrates, as a rule, has a high phosphorus content, which limits the use of medium-carbon ferromanganese in steelmaking. Therefore, to obtain medium-carbon ferromanganese with a regulated by standard low (0.1% P) and decreased (<0.4% P) phosphorus content, it is necessary to use high-carbon ferromanganese smelted with low-phosphorous manganese ore (imported) or low-phosphorous manganese charge slag, that significantly increases the cost of initial ferromanganese and, as a result, medium-carbon ferromanganese.

Among other requirements for the initial high-carbon ferromanganese, the low silicon content is of primary importance. In the process of blowing the melt, silicon is oxidized in the first minutes of ferromanganese conversion. The resulting silica interacts with manganese oxide, increases the chemical aggressiveness of the slag melt in relation to the

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periclase lining. Acidic slag, interacting with basic lining, reduces its stability, thus necessitating the use expensive highly refractory periclase refractories, including electromelted magnesite (periclase).

The increased silicon content in the initial high-carbon ferromanganese increases the amount of SiO<sub>2</sub> in the slag, that is accompanied by a high consumption of lime, which should also have a low silica content to reduce the slag ratio and the loss of manganese with waste slag. To reduce metal loss at the gas phase, the oxidative conversion of low-silicon high-carbon ferromanganese for carbon oxidation must be carried out under a rational temperature regime. **The aim** of the work is to establish physicochemical patterns of behavior of carbon, silicon, manganese when using the method of oxygen purge of high-carbon ferromanganese

### Theoretical studies of decarburization of a high-carbon ferromanganese melt.

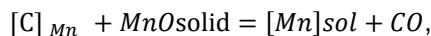
In solid high-carbon initial ferromanganese (~ 8% C), carbon is represented by carbide [Mn, Fe]<sub>7</sub>C<sub>3</sub>. In liquid manganese, carbon is dissolved in a significant amount. At the temperature of 1773K, the solubility of carbon in liquid manganese is 7.892%, and at 1873K it is 8.116%. [1] When producing a number of steels

$$\ln\gamma_{[C]} = (1 - x^2) \cdot [-0.745 - 0.97x - 55.9x^2 + 771x^3 - 1432x^4]$$

$$\ln\gamma_{[Mn]} = x^2 [4.5 + 85x - 904x^2 + 1531x^3]$$

In studies, liquid manganese and graphite have been taken as the standard state [3]. In the calculations, the values of saturated solutions of carbon in manganese can be taken as the standard state [4].

The study of the activity of manganese and carbon by the equilibrium method of the Mn-C system melt with solid MnO oxide and the gas phase of Ar-CO with controlled partial pressure of CO by the reaction [5],



$$K_p = \frac{a_{Mn} \cdot P_{CO}}{a_{[C]Mn} \cdot a_{MnO}}$$

has established the equilibrium constant of the above reaction and the concentration dependences  $a_{Mn}$   $a_c$  [5]. The dependences of manganese and carbon activity coefficients on the manganese and carbon molar fractions in the alloy are described by the equation:

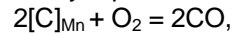
$$\ln\gamma_{Mn} = 5,47x^2 - 46,0x^3$$

$$\ln\gamma_C = -0,291 - 10,9x + 75x^2 - 46,8x^3$$

Oxidative refining of high-carbon ferromanganese in an oxygen bottom-blown converter has shown [6] that the ratio [% Mn] / [% C] depends on temperature, and this dependence is satisfactorily described by the equation:

and alloys, it is necessary to have ferromanganese which has low carbon content. Currently, medium- and low-carbon ferromanganese are obtained by silicon reduction of manganese oxides due to silicon of ferrosilicon manganese [2].

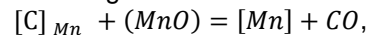
The process of decarburization of liquid ferromanganese with gaseous oxygen can be generally represented by the reaction:



$$K_{O_2} = \frac{P_{CO}^2}{a_c^2 \cdot P_{O_2}},$$

$$\text{where } a_c = \gamma_c X_{[C]}$$

In the case of oxidation of carbon dissolved in manganese, the reaction of its interaction with MnO is the following:



$$K_{MnO} = \frac{a_{[Mn]} \cdot P_{CO}}{a_{[C]} \cdot a_{(MnO)}},$$

$$a_{[Mn]} = \gamma_{[Mn]} X_{[Mn]}.$$

The calculation of the activity coefficients  $\gamma_{[C]}$  and  $\gamma_{[Mn]}$  depending on the silicon content in ferromanganese can be carried out according to the equations [3]:

$$\lg \frac{[\%Mn]}{[\%C]} = -\frac{10115}{T} + 6,70$$

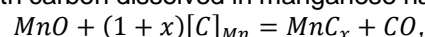
With increasing temperature, the carbon content decreases, but at the same time, the manganese vapor pressure also increases significantly, that follows from the equation

$$\lg P_{Mn} = -\frac{12546}{T} + 10,483 \text{ (Па)}$$

The decarburization of ferromanganese with gaseous oxygen should be carried out at relatively moderate temperatures to reduce the loss of manganese due to evaporation and the rate of carbon oxidation with the reduction of manganese loss due to evaporation should be matched.

Refining of high-carbon ferromanganese with manganese ore and high manganese slag melts was carried out by pioneering studies of V.A. Boholiubov [7.8]. High-carbon ferromanganese mixed with manganese ore was melted in an electric steel furnace. The chemistry of the refining process is quite simple, and does not require the production of ferrosilicon manganese, which is attractive for enterprises that smelt ferromanganese only.

According to N.V. Tolstoguzov [9], the change in the Gibbs energy of the interaction reaction of MnO with carbon dissolved in manganese has the form:



$$\Delta G_T^\circ = 196293 - 123.0 \cdot T, \text{ J/mol.}$$

The theoretical equilibrium temperature of this reaction, taken as the reaction initiation temperature, is 1597K (1324 °C).

The authors [10] explain the difficulties in obtaining refined high-carbon ferromanganese by oxidative refining due to the formation of a carbon solution in manganese during the interaction of liquid manganese with carbon monoxide.

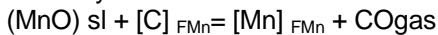
As shown in the paper [11], the interaction reaction of carbon from high-carbon ferromanganese with manganese oxide can be appreciably developed even at a temperature of 1600 °C, and a content of 2% can be achieved at a temperature of about 1750°C. Due to the tense heat balance, refining of high-carbon ferromanganese with air did not receive industrial implementation. However, the ideas laid down in experiments for blowing ferromanganese melt with air were developed in subsequent works on decarburization of ferromanganese with gaseous oxygen.

**The results of experimental studies. Decarburization of high-carbon ferromanganese with oxygen in a ladle and in an induction furnace**

When refining [12] ferromanganese in IST-0.16 furnace and in a ladle, gaseous oxygen has been blown. During the blowing the melt, the dependence of the change in the ratio [%C]/[%Mn], which depends on temperature, was established. The process of decarburization of ferromanganese was conventionally divided into three periods.

In the first period, silicon and manganese were oxidized by top-blown gaseous oxygen supplied to the ferromanganese melt, which was accompanied by the formation of slag and the intense evolution of red fume, consisting of dusty particles of manganese oxides. As a result of the exothermicity of silicon and manganese oxidation reactions, the melt temperature increased to 1540–1560°C. However, the carbon content in the ferromanganese melt practically did not change. Only at the end of the first period, the flame from CO burning appear, it was considered a sign of the start of carbon burning.

In the second period, carbon began to oxidize intensively and the flame from CO burning increased. Manganese oxide of the slag was spent on carbon oxidation by the reaction



and until the end of the second period, the MnO content decreased markedly. The temperature of the metal increased to 1680-1720°C.

In the third period, the co-oxidation of carbon and manganese took place. The CO flame decreased and the amount of red fume increased. The temperature increased to 1750-1820°C.

During the blowing, the ratio [%C]/[%Mn] remained constant up to 1540-1560°C, which indicates the practical absence of decarburization of high-carbon

ferromanganese. An increase in the temperature of the ferromanganese melt occurred due to the further oxidation of manganese. The increase in temperature was accompanied by a significant decrease in the ratio [%C]/[%Mn], which indicated the intensive development of the decarburization of ferromanganese. At 1700–1800°C, the ratio [%C]/[%Mn] changed insignificantly; therefore, the decarburization reaction of ferromanganese in the industrial amount faded out and mainly manganese continued to be oxidized [11]. The authors [11] has generalized the results of research melts on high-carbon ferromanganese refining from carbon by top-blown oxygen and has shown that the manganese content in the alloy increased with decreasing carbon concentration. The phosphorus fraction increased by 0.05-0.1%. Large losses of manganese during the smelting of manganese alloys are caused, as noted above, by high vapor pressure - 13.33 kPa at 1560°C and 66.65 kPa at 1820°C [13.2].

When refining ferromanganese by oxygen blowing, up to 10% Mn is oxidized and lost with dust (% by mass) in which manganese is contained in the form of Mn<sub>2</sub>O<sub>3</sub>:

Mn <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	MgO	FeO	P
94-96	1,0	1,5	1,5-2,0	0,03-0,05

The use of high-carbon ferromanganese with a low silicon content (less than 1%) as the initial alloy for decarburization is associated with a deterioration in the heat balance and additional losses due to manganese oxidation and its loss with slag. Therefore, high-carbon ferromanganese in experiments for oxygen blowing is used with a silicon content of (≥ 2.0 - 2.5%).

In order to clarify the specifics of high-carbon ferromanganese decarburization (with a content of 2.5–4.0% Si) when blown with oxygen [12], it was established that at blowing silicon carbon ferromanganese with oxygen, silicon is oxidized primarily, manganese is oxidized simultaneously, but to a lesser extent. In this case, despite a significant increase in temperature to 1650-1680 °C, the decarburization reaction did not receive significant development. Intensive decarburization of ferromanganese began with a residual silicon content of 0.2-0.5%. By the end of the blow, the content of silicon and phosphorus increased due to loss of alloy mass associated with the burn-out of carbon and manganese.

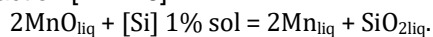
To reduce the activity of silica in the slag, lime was added in portions into the furnace during the blowing. Lime had a beneficial effect on the decarburization of ferromanganese - melting was performed smoothly, without emissions, the blowing mode was stabilized, and the rate of carbon oxidation increased (on average by 0.02 - 0.05% C per minute). The final slag of the heats had the following chemical composition,%:

Heat	MnO	SiO <sub>2</sub>	CaO	MgO	FeO	C	P	%CaO/%SiO <sub>2</sub>
4	50,0	14,72	23,5	7,72	1,78	0,08	0,064	1,59
5	45,1	18,52	24,3	7,00	1,52	0,04	0,045	1,31

During the oxidative decarburization of high-carbon ferromanganese, the fractional concentration of phosphorus does not decrease, but rather increases because phosphorus has significantly lower chemical affinity for oxygen compared to manganese. Due to the physicochemical features of phosphorus itself - and its solutions in manganese (or in melts containing iron), the initial high-carbon ferro manganese for oxidative refining should have a lower phosphorus content than standard, since the allowed phosphorus content in medium-carbon ferromanganese regulated by the standard should not exceed 0.4%.

#### **Thermodynamic prerequisites for the interaction of manganese protoxide and silicon.**

The process of reducing manganese from pure liquid manganese protoxide by silicon dissolved in manganese can be represented by the chemical reaction [14 - 16]



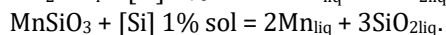
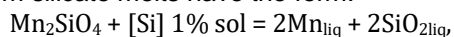
The temperature dependence of the Gibbs energy of the reaction, taking into account the thermodynamic data of the phase transformations of the intermediate reactions, has the form

$$\Delta G_T^0 = -98680 - 111,94 \cdot T + 13,55 \lg T - 19,85 \cdot 10^{-3} T^2, \text{ Дж г-ат Si.}$$

The approximation of the functional dependence for the temperature range 1250-1700 ° C leads to a simplified record of the equation:

$$\Delta G_T^0 = -47000 + 91,45 \cdot T$$

For more complete thermodynamic analysis of the silicothermal process, it should be taken into account that the reduction of manganese for metallic manganese and medium-carbon ferromanganese does not occur from pure manganese protoxide, but from manganese-silicate melts, for which one can accept the existence of both  $\text{Mn}_2\text{SiO}_4$  (an analogue of the natural mineral tephroite) and  $\text{MnSiO}_3$  (rhodonite). Reactions for the reduction of manganese from silicate melts have the form:



The introduction of calcium oxide into the system  $\text{MnO-SiO}_2$  melt increases the thermodynamic conditions for a more complete manganese reduction due to an increase of manganese protoxide activity and a decrease of silicon activity in the slag. Incomplete reaction to equilibrium low concentrations of  $\text{MnO}$  in the slag during the process is due to both a decrease in the activity of manganese protoxide and the activity of silicon dissolved in manganese.

**Scientific novelty.** Taking into consideration the high affinity of silicon for oxygen, the physical and chemical basis for the production of medium-carbon ferromanganese, as well as metallic manganese and low-carbon ferromanganese, is the process of the interaction of manganese oxides of a certain basicity slag melt with silicon dissolved in ferromanganese (manganese), that is, as combined reduction-refining process to produce manganese ferroalloys with a given silicon content standard.

The technology of silicothermal reduction of manganese oxides from slag was tested in a bottom-blown converter. The experimental melts were carried out in a 15-ton converter with three bottom tuyeres of the pipe-in-pipe type made of copper and stainless pipes. Converter is lined with periclase-chromite refractory material. To melt MnC17 grade silicomanganese, an arc furnace was used, which was also lined with periclase-chromite brick. After the MnC17 was melted and heated to temperatures equal to 1620–1630°C, the melt was poured into the converter with the help of a transfer ladle. Freshly calcined lime in an amount of 20 kg / t was added to the hot converter, on its hearth, before liquid silicomanganese was poured off.

For 5-6 minutes, in order to heat the metal bath, oxygen was injected into the converter at intensity of 0.98-1.02 m<sup>3</sup>/t·min. together with natural gas, intensity of which was of 7-10% of the oxygen consumption. During this period, the metal was heated to temperatures of 1670-1680°C. Then argon blowing was introduced, supplied both by the central tuyere channels and by the peripheral ones with a total intensity of 0.7 m<sup>3</sup>/t · min. When switching to blowing the converter bath with neutral gas, the fluxed agglomerate of the following composition: MP - 33.9%; SiO<sub>2</sub> - 14.8%; CaO - 28.3% was added to the converter. During this period, the temperature of the metal bath of the converter decreased to 1320-1340°C.

Then, to preheat the melt, a 6-7 minute oxygen blowing was used. During this period, the metal was heated to a temperature of 1600-1620°C. After preheating the alloy, neutral gas blowing was applied again. Freshly calcined lime and fluxed agglomerate were again added to the converter. Then neutral gas was blown for 7-9 minutes. At the same time, the metal temperature was decreasing to 1300-1320 ° C. The output of the melt was carried out to a mould lined with chromomagnesite brick through the mouth of the converter.

In general, the refining process in the bottom-blown converter took place calmly, without any emissions and slashings. Slag throughout the blowing was rarely unstable, and its basicity ((CaO MgO) / SiO<sub>2</sub>) at the end of the refining period was not less than 1.2-1.3. The content of silicon in the metal in the ladle was 1.60-1.38%.

#### **Conclusions**

When melt is heated by oxygen blowing, predominant oxidation of silicon occurs. Its presence in the metal practically blocks the oxidation of manganese. Since oxygen is an assimilating gas, the processes of mixing the components of the converter bath and the reduction of manganese oxides at the metal-slag interface do not receive appropriate development during blowing.

Heats of medium-carbon ferromanganese in a bottom-blown converter are characterized by a stable chemical composition and they almost completely meet the requirements for this type of alloy. A slight

excess of the concentration of silicon in the metal on a number of heats can be easily eliminated by additional blowing of the bath with oxygen at the final stage of refining.

The behavior of phosphorus in these heats has not controlled. The content of P<sub>2</sub>O<sub>5</sub> in the final slag is 0.1%. To achieve acceptable concentrations of phosphorus in the metal, it is necessary to use low-phosphorous initial charge materials.

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