Basics of Hybrid Energy-Metallurgical and Catalytic Technology of Synthetic Methane Production

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Abstract

In this paper, we considered the approach to reducing the level of GHGs emissions below the limit, which the reduction reaction of iron oxides by carbon determines. We have shown this is possible if a part of the gases (CO) leaving the metallurgical unit will be processed into synthetic methane or other commercial chemical products. For this purpose the metallurgical unit of continuous action is necessary. We looked at the physical-chemical basis of operation of such a unit on the basis of smelting reduction (SR) in an electric arc furnace. In this unit, a mixture of powders of stoichiometric composition from iron ore magnetite concentrate and coal is fed through the tuyeres deepened to the bottom of the metal bath by injection (CO is the injecting gas). These tuyeres are located under the electrodes. As a result, a reaction zone occurs in a metal bath (low carbon steel) in each cylindrical region that extends from the tuyere to the electrode. In this reaction zone, FeO dissolves in a metal bath (without slag formation) and is reduced by solid carbon particles, which float up. The mathematical model of this process is created. For the reaction zone under consideration, an equation for calculating the productivity (relative to the mass of reduced iron) was obtained. The calculation results show that a metallurgical unit with 6 reaction zones (6 electrodes in an electric arc installation) with a total volume of a metal bath (including all 6 reaction zones) of about 16 m^3 has a design capacity of about 411 t/h of an iron-carbon product (low-carbon steel with a carbon content of about 0.2 %). For example, such a productivity corresponds, to the total performance (by weight of blast furnace iron) of a blast furnace shop, which has several blast furnaces to a total volume of about 7000 m^3. The considered process of hybrid energy-metallurgical and catalytic technology of synthetic methane production is capable to replace equally (regarding overall productivity) blast-furnace and converter production, and also, about these productions, to provide double reduction of technogenic loading on environment.
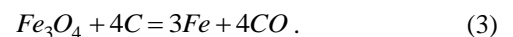
Keywords: SR process; synthetic methane; greenhouse gases; SR process model; emerging technology for steelmaking

I. INTRODUCTION

The process of direct reduction of iron oxides (magnetite) by carbon is generally characterized by the following reactions [1]:
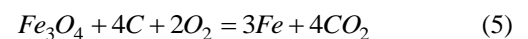
$$Fe_3O_4 + C = 3FeO + CO, \tag{1}$$

$$FeO + C = Fe + CO. \tag{2}$$
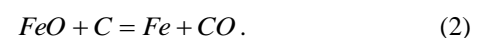
In sum, the reactions (1) – (2) can be represented as:

$$Fe_3O_4 + 4C = 3Fe + 4CO. \tag{3}$$

The carbon monoxide produced by the reaction (3) is afterburned either in a metallurgical unit or in technical devices that use CO as a fuel:

$$2CO + O_2 = 2CO_2 \tag{4}$$
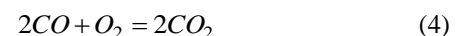
The chemical processes of reduction of iron (3) and afterburning of CO (4) we present summarily as:

$$Fe_3O_4 + 4C + 2O_2 = 3Fe + 4CO_2 \tag{5}$$

When using coal as a reducing agent, CO_2 emissions in modern metallurgical technologies cannot be reduced below the limit set by the reaction (5). To overcome this limitation,

new technologies must be created. In this regard, as a component of these technologies, the process of smelting reduction (SR) is of significant interest.
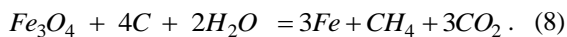
In the world practice of scientific research SR process, as an alternative to the domain process, has been considered repeatedly. It is known that various variants of this process allow to produce both cast iron and steel [2]. It has been shown experimentally that it is possible to obtain both medium-carbon steel [3] and low-carbon steel [4] using this process.

When implementing this process, the waste gases (a mixture of CO and CO_2) can contain from 60 to 90 % CO [4]. This creates prerequisites for the chemical catalytic processing of this gas mixture, for example, into synthetic methane or other chemical products.

The process of producing low-carbon steel in an electric arc furnace (based on the SR process) and the processes of catalytic production of methane from CO are characterized by a number of chemical reactions. The reaction (3) is carried out in a metallurgical unit, and chemical reactions take place in a system of chemical reactors that produce synthetic methane [5]:

$$CO + H_2O = H_2 + CO_2 ; \qquad (6)$$

$$CO + 3H_2 = CH_4 + H_2O ; \qquad (7)$$
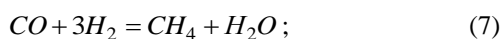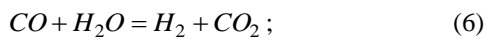
We can sum up the reactions (3), (6) and (7) as:

$$Fe_3O_4 + 4C + 2H_2O = 3Fe + CH_4 + 3CO_2 . \quad (8)$$

The chemical processes that reaction (8) reflects are the chemical processes of a hybrid energy-metallurgical and catalytic technology of synthetic methane production. From the comparison of equations (5) and (8), we see that CO_2 emissions in the hybrid energy-metallurgical and catalytic technology of synthetic methane production are 25% less compared to existing methods of carbon-thermal reduction of iron (processes that are reflected by the reaction (5)). Taking into account that there is no combustion of coal (metallurgical fuel) in the process analyzed by SR, the application of technological processes reflected by the reaction (8) will ensure a reduction in greenhouse gas emissions relative to the total emissions of blast furnace and converter production by approximately 50%.

Experimental results [6] show that reduction of iron from oxide (iron oxide dissolved in iron melt) by carbon proceeds at a rate of $(1,1 - 3)10^{-4}$ mole-$FeO / (cm^2 \ sec)$ at $1693 - 1893$ K. This is approximately 5 times greater than the rate of reduction of iron from oxides by solid carbon also at $1693 - 1893$ K in the slag melt. For the stated reason, we consider the metallurgical unit of hybrid energy-metallurgical and catalytic technology of synthetic methane production as a equipment of continuous operation with electric arc heating of a metal bath. This unit should provide a content of about 0.2% carbon (the rest-iron) in the metal discharged. This unit must have a number of tuyeres deepened into metal bath, which inject the following materials into the melt:

– coal powder (carburizer), which is fed only as needed to maintain a constant concentration of carbon (about 0.2%) in the iron-carbon melt drained from the metallurgical unit. As an injection gas, we consider the use of CO;

– a mixture of magnetite (iron ore concentrate) and coal powder (the mixture has stoichiometric composition and contains carbon, necessary only for the reduction of iron, i.e. 0.207 kg of carbon per 1 kg Fe_3O_4). As the gas providing injection of this mixture into a metal bath, we assume the use of CO.

II. RESEARCH METHODOLOGY

A. *Physical-chemical processes in a metal bath during injection of a powdered mixture of iron oxide and carbon*

We view a tuyere deepened to the bottom of the metal bath to inject a mixture of magnetite iron ore concentrate and coal powder. Since the components of the mixture float up after injection, then, in this case, the reaction zone **A** is a column of melt over the tuyere (see Fig. 1).
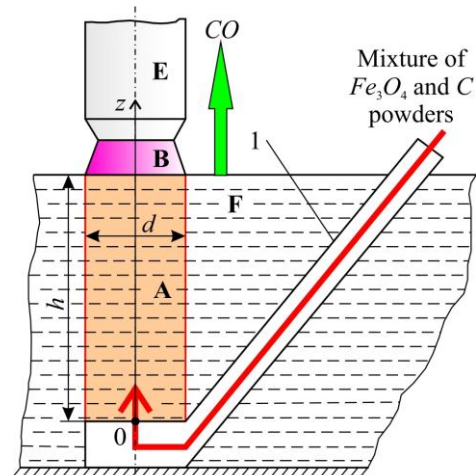

Fig. 1. Arrangement of reaction zone **A** in the metal bath **F** above the injection tuyere 1: **B**-electric arc; **E** -electrode; h, d -height and diameter of the zone **A**; z -coordinate with zero mark at the level of the tuyere

For modern electric arc steel furnaces, the depth of the metal bath can reach 1.3 m, and the diameter of the electrodes can be up to 0.61 m [7]. We therefore adopted the parameters of the reaction zone **A** (see Fig. 1) $h \approx 1$ м, $d \approx 0,6$ м.

The results of modeling the temperature field in the near-electrode region of a metal bath in an arc steelmaking furnace show that the temperature T of the metal bath under the electrode varies from 3000 K on its surface to 2000 K at a depth of 1 m [8, 9].

The heating time of the injected powders to the melt temperature is negligible. For example, for coal particles with a size of 2.3 mm, the heating time from room temperature to 1700 K is less than 1 s at typical ascent rates [10].

Magnetite when heated in nonoxidation gas is transformed into wustite by reaction [11]:

$$2Fe_3O_4 \leftrightarrow 6FeO + O_2 . \qquad (9)$$
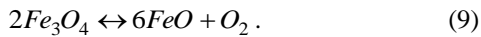
Experimental studies have shown that reaction (9) proceeds efficiently at $T > 1773\,K$ [11]. Consequently, powdered Fe_3O_4 injected into reaction zone **A** (see figure 1) will turn into FeO during heating to reaction zone temperature. This means that the injected iron oxides dissolve in the metal bath by reaction:

$$FeO \leftrightarrow [FeO]. \qquad (10)$$

A number of authors proposed several mathematical models for the dissolution of coal powder in iron melt, which are confirmed by the results of experimental studies [11-13]. However, these studies were performed in relation to oxygen-free iron-carbon alloys. At the same time, it is known that when a carburizer (coal powder) is added to the steel melt, it is covered with gas bubbles (CO) and only after the oxygen concentration in the metal bath decreases to approximately 0.02% in the contact zone with the carburizer, the dissolution of carbon in the steel melt begins [14].

Observations in x-rays of the processes of occurrence and formation of gas bubbles in the interaction of solid carbon with iron oxide melt show the existence of two interfaces: gas-melt and gas-solid carbon, i.e. solid carbon does not directly contact the melt during the reduction of iron oxides [15 – 17].

In so doing, a chemical reaction occurs on the gas-melt interface [17]:

$$[FeO] + CO \leftrightarrow [Fe] + CO_2 \qquad (11)$$

and on the surface of the gas-solid carbon interface regeneration of the reducing agent occurs by reaction:

$$C + CO_2 \leftrightarrow 2CO . \qquad (12)$$

The general record of reactions (11) and (12) is as follows:

$$[FeO] + C \leftrightarrow [Fe] + CO \qquad (13)$$

Thus, in the reaction zone **A** of the considered metallurgical unit, the following reactions occur: dissolution of iron oxides in the melt (10) and reactions (11) and (12), which provide reduction of oxides dissolved in the metal bath by carbon. We calculated the equilibrium constants of these reactions according to the data [18]. These constants, expressed in terms of reagent concentrations, have the following form. For reaction (11) it is:

$$\frac{[CO_2]}{[CO]} = 10^{\frac{11824,.8}{T} - 6,982} \qquad (14)$$

and for reaction (12) it is:

$$\frac{[CO]^2}{[CO_2]} = 10^{\frac{-9009,.8}{T} + 9,2198} . \qquad (15)$$

It follows from equation (14) that in the reaction zone **A** (see Fig. 1), gas bubbles (they occur around the grains of powder

of carbon) is dominated by *CO*. As established in [16, 19, 20], the reaction (12) is not limiting. The rate of the reduction process (13) is determined by the reaction (11), which proceeds in the diffusion mode, and the limiting stage of this reaction is the diffusion of oxygen to the gas-melt interface [16, 20].

B. Model

Given that when a gas bubble up floats $Pe = \dfrac{wr_g}{D} \gg 1$ (w_g is the speed of up floating bubble; r_g is the radius of a gas bubble that up floats without crushing; D is diffusion coefficient of oxygen in the melt), then near the surface of the bubble, a diffusion layer arises. The distribution of oxygen concentration (c in equation (16)) in the boundary layer is described by the convective diffusion equation, which in spherical coordinates (r, θ, because the concentration φ does not depend on the azimuth) have the form [21]:

$$v_r \frac{\partial c}{\partial r} + v_\theta \frac{1}{r} \frac{\partial c}{\partial \theta} = D \frac{\partial^2 c}{\partial r^2} \qquad (16)$$

where v is the velocity of the fluid in the boundary layer.

The initial and boundary conditions for the carbon particle injected into the melt are as follows:

$$\begin{cases} 1,5w = v_0 & \text{by } t = t_0 \\ c = c_1 & \text{by } r = r_g \\ c = c_0 & \text{by } r \to \infty \end{cases} \qquad (17)$$

where v_0 is the velocity of the liquid at the surface of the bubble; c_0 – is the concentration of oxygen in the melt; c_1 is the concentration of oxygen at the gas-melt interface. As stated above $c_1 \approx 0,02\%$.

V. G. Levich [21] obtained the solution of equations (16) – (18). Using this solution, the expression for the total diffusion flow of oxygen from the melt to the surface of the up float gas bubble is (kg / s):

$$I_O = 8(c_0 - 0,02)10^{-2}\rho_L \sqrt{\frac{\pi}{2}} \left(\frac{Dw_g}{r_g} \right)^{1/2} r_g^2 \qquad (18)$$

where ρ_L is the density of the melt. Equation (18) using the equations below can be converted into an equation for calculating the performance (by weight of reduced iron) metallurgical unit of hybrid energy-metallurgical and catalytic technology of synthetic methane production.

The radius of the gas bubble that up float without crushing is determined by the equation [22] (mm):

$$r_g = 0,72\sigma_m^{1/2}\rho_L^{-1/6}\rho_g^{-1/3}g^{-1/2} \qquad (19)$$

where σ_L is the surface tension of the melt; ρ_g is the density of the gas in the bubble; g is the acceleration of free fall. The characteristic radius of the bubbles formed is about 5 mm.

The surface tension of the iron melt in the temperature range from 1843 K to 2150 K is determined by the dependence [23] (N·m^{-1}):

$$\sigma_L = 1{,}978 - 4{,}9\left(T - 1843\right). \qquad (20)$$

We used this dependence as a first approximation for the temperature interval in the reaction zone **A** (see Fig. 1).

The density of the iron-carbon melt with a carbon content of 0.2% in the temperature range from 2000 K to 3000 K is determined by the dependence that we obtained on the basis of data [23] (kg/m^3):

$$\rho_L = 6970 - 0{,}68\left(T - 2000\right). \qquad (21)$$

Given that the density of CO at a pressure of 1 bar and a temperature of 2000 K is $\rho_0 = 0{,}16841$ kg/m^3 [24], the following equation follows from the gas state law (kg/m^3):

$$\rho_g = 2 \cdot 10^{-2} \rho_0 \frac{P_g}{T} \qquad (22)$$

where the pressure in the gas bubble P_g corresponds to the sum of the atmospheric pressure P_a and the pressure of the melt column, which depends on the z coordinate (see Fig. 1) (Pa):

$$P_g = P_a + \rho_L g\left(1 - z\right). \qquad (23)$$

The rate of bubble ascent is determined by the equation [21] (m/s):

$$w_g = \left(\frac{4g\sigma_L^2 \rho_g}{\alpha \mu_L \rho_L^2}\right)^{1/5} \qquad (24)$$

where $\alpha \approx 30$ is the empirical coefficient [21]; μ_L is the dynamic viscosity of the melt. For μ_L in the considered temperature range the dependence [25] is used (Pa·sec):

$$\mu_L = 10^{\left(\frac{2385{,}2}{T} - 3{,}5958\right)}. \qquad (25)$$

The characteristic rate of ascent of gas bubbles is about 0.1 m/s.

The ascent rate of coal particle is determined by the Stokes law [21] (m/s):

$$w_C = \frac{2}{9} \frac{\left(\rho_L - \rho_C\right)g R_C^2}{\mu_L} \qquad (26)$$

where $\rho_C = 1600$ kg/m^3 is the density of coal (anthracite) [26]. Under the conditions under consideration, the rate of ascent of coal particles having a radius of $R_C = 0{,}5 \cdot 10^{-3}$ m is about 1 m/s.

The concentration of oxygen in the melt, at which no slag FeO is formed in the temperature range from 1880 K to 2230 K, is [26] (wt. %):

$$[O] = 10^{\frac{-6380}{T} + 2{,}765}. \qquad (27)$$

We use the dependence (27) in the temperature range of the reaction zone **A** (see Fig. 1).

The temperature dependence of the oxygen diffusion coefficient in liquid iron at temperatures from 1833 K to 1933 K according to [27] has the form (m^2/s):

$$D = 5{,}59 \cdot 10^{-7} exp\left(\frac{-19500}{RT}\right) \qquad (28)$$

where R is the universal gas constant. Equation (28) in the first approximation we used in the temperature range from 2000 K to 3000 K.

The number of gas bubbles in the reaction zone in which the reactions (11), (12) occur is determined by the following equation:

$$N = \frac{3d^2 h \varphi_g}{16 r_g^3} \qquad (29)$$

where φ_g is gas content.

Taking into account the above equations (19) – (29) and stoichiometric relations in the reaction (13), we have transformed equation (18) into equation (30), which determines the reaction zone **A** output (weight of reduced iron, kg/s):

$$I_{rz} = 8\left(c_0 - 0{,}02\right)10^{-2} \rho_L \sqrt{\frac{\pi}{2}} \left(\frac{Dw_g}{r_g}\right)^{1/2} r_g^2 \frac{M_{FeO}}{M_O} N \qquad (30)$$

where M_{FeO}, M_O are the molar masses FeO and O, respectively.

III. RESULTS AND DISCUSSION

From the above data, we see that coal particles float up much faster than gas bubbles ($w_C \gg w_g$). Consequently, gas bubbles are constantly formed around the coal particle when it rises. This allows us to conditionally consider the process of reactions (11) and (12) in the system: a spherical carbon particle and the surrounding it gas bubble that is in contact with the melt. In this case, the mass consumption of carbon for the reduction of iron from the oxide is (kg/s):

$$I_C = I_O \frac{M_C}{M_O} \qquad (31)$$

where M_C is molar mass of carbon.

The calculation according to equation (31) shows that the carbon particle with $R_C = 0{,}5 \cdot 10^{-3}$ m will completely disappear as a result of chemical reactions (11) and (12) for 0.02 s. At the initial rate of ascent of the carbon particle $w_C = 1{,}2$ m/s, the height of its ascent, at which it will cease to exist, is 0.024 m, while the mass concentration of oxygen on the surface of the gas-melt bubble surrounding the particle will decrease to 0.02%. This means that other carbon particles will upfloat to a great distance, where they will fully react

according to the generalized reaction (13), and so on, sequentially, along the entire height h of the reaction zone **A**.

The solution of equation (30) in $\varphi_g = 0.2$ gives the value of the total productivity of the reaction zone **A** (see Fig. 1) for reduced iron $I_{rz} = 19$ kg/s.

This means that, for example, for the 6 reaction zones located in the metallurgical unit of the hybrid energy-metallurgical and catalytic synthetic methane production technology (see reaction (8)), which are arranged as shown in the cross section of the metal bath in Fig. 2, the total capacity of this metallurgical unit will be 411 t/h of iron-carbon product (low-carbon steel with a carbon content of 0.2 %). For example, such a productivity corresponds to the total performance (by weight of blast furnace iron) of a blast furnace shop, which has several blast furnaces to a total volume of about 7000 m³ that is about 460 t/h [27].
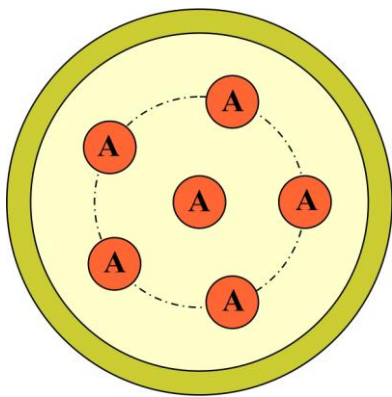

Fig. 2. Approximate arrangement of 6 reaction zones along the cross section of the metal bath of the metallurgical unit under consideration

IV. CONCLUSION

In this paper, we have outlined the physical-chemical basis of hybrid energy-metallurgical and catalytic technology of synthetic methane production (see reaction (8)), considered the main elements of the mathematical model of this technology and conducted numerical studies. The results show that:

- Reduction in greenhouse gas emissions below the limit level (this level is determined by the chemical reaction of reduction of iron from oxides by coal (5)) is feasible if new hybrid chemical-metallurgical technologies are used, in which part of the gases leaving the metallurgical unit is processed into commercial chemical products.

- The considered process of hybrid energy-metallurgical and catalytic technology of synthetic methane production is capable to replace equally (regarding overall productivity) blast-furnace and converter production, and also, about these productions, to provide double reduction of technogenic loading on environment.

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