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Oxidation of Molten Impurities in Converters by Means of Combustion Flames: Thermodynamic Principles. 1. Thermodynamic Analysis of Processes in Natural-Gas Combustion

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Abstract—In the production of steel, as the productivity rises and the resource and energy consumption declines, improvements in converter design are required to ensure preliminary scrap and batch heating and to intensify redox processes in the liquid bath and exhaust-gas combustion above the bath, without impairing the durability of the injection systems and the converter lining. The use of fuel–oxygen combustion flames in the converter resolves numerous technological problems. The hydrodynamics in the reaction zones and in the liquid bath may be greatly changed by fuel combustion in the converter's working space with jet formation or by means of submersible combustion flames. In the present work, thermodynamic methods are used to analyze the dynamics of gaseous-fuel combustion and the oxidation of elements in the converter bath on interaction with high-temperature combustion products. The interaction of the combustion flame and chemical elements in the converter bath is calculated for equilibrium conditions. The use of the combustion flames is found to change the composition of the gas phase in the converter's working space (above the bath), which contains H_2 and H_2O in addition to the traditional components associated with oxygen injection: O_2, CO, CO_2. The presence of H_2 and H_2O changes the thermal conditions and oxidative properties of the gas phase. In the combustion of gas–oxygen fuel, the optimal composition of the initial gas mixture (natural gas + oxygen) must correspond to the ratio 100% CH_4 + 69% O_2. The oxidation product is gaseous phase consisting of 40% CO_2 + 60% H_2O. The total enthalpy of combustion of the gas–oxygen fuel at converter temperatures, with an oxygen excess greater than 1.0 (up to 2.0), is about 200 kJ per mole of the initial reagents. In the oxidation of methane by carbon dioxide, the total enthalpy of combustion is between −7 and −14.5 kJ/mol of initial reagents at 1800 K. The process becomes endothermal at temperatures above 2000 K: $\Delta H_{2200} = 7.7–15.4$ kJ/mol. In the oxidation of natural gas by water vapor, $\Delta H_{1800–2200} = 19.5–70$ kJ/mol. Thus, flame temperatures above 1800 K may only be attained in the oxidation of methane by oxygen. The use of air, carbon dioxide, or water vapor as the oxidant does not yield the required thermal effect.

Keywords: converter, gas–oxygen combustion flames, methane, hydrocarbons, steel, oxygen injection, thermodynamic analysis

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As we know, steel production in the oxygen converter was originally developed as a simple process, without any added fuel [1–12]. In the classical approach, as a rule, the quantity of hot metal, its composition, and the temperature determine the oxygen consumption in the process and the quantity of cooling material required to absorb the excess heat. In most cases, metal scrap is primarily used for cooling.

In recent years, the basic approaches to determining the optimal smelting conditions in the converter have changed significantly, on account of the introduction of resource- and energy-saving technologies and the expansion of waste processing. The introduction of complex (top and bottom) injection of the converter bath has significantly expanded the functional capabilities of the system in terms of cooling by means of sinter, pellets, rich manganese ores, and so on [1]. That increases the yield of molten steel and considerably reduces the combustion of manganese-bearing ferroalloys. In Japan, for example, around 50% of the manganese and up to 15% of the chromium in the steel is supplied to the converter in the form of relatively

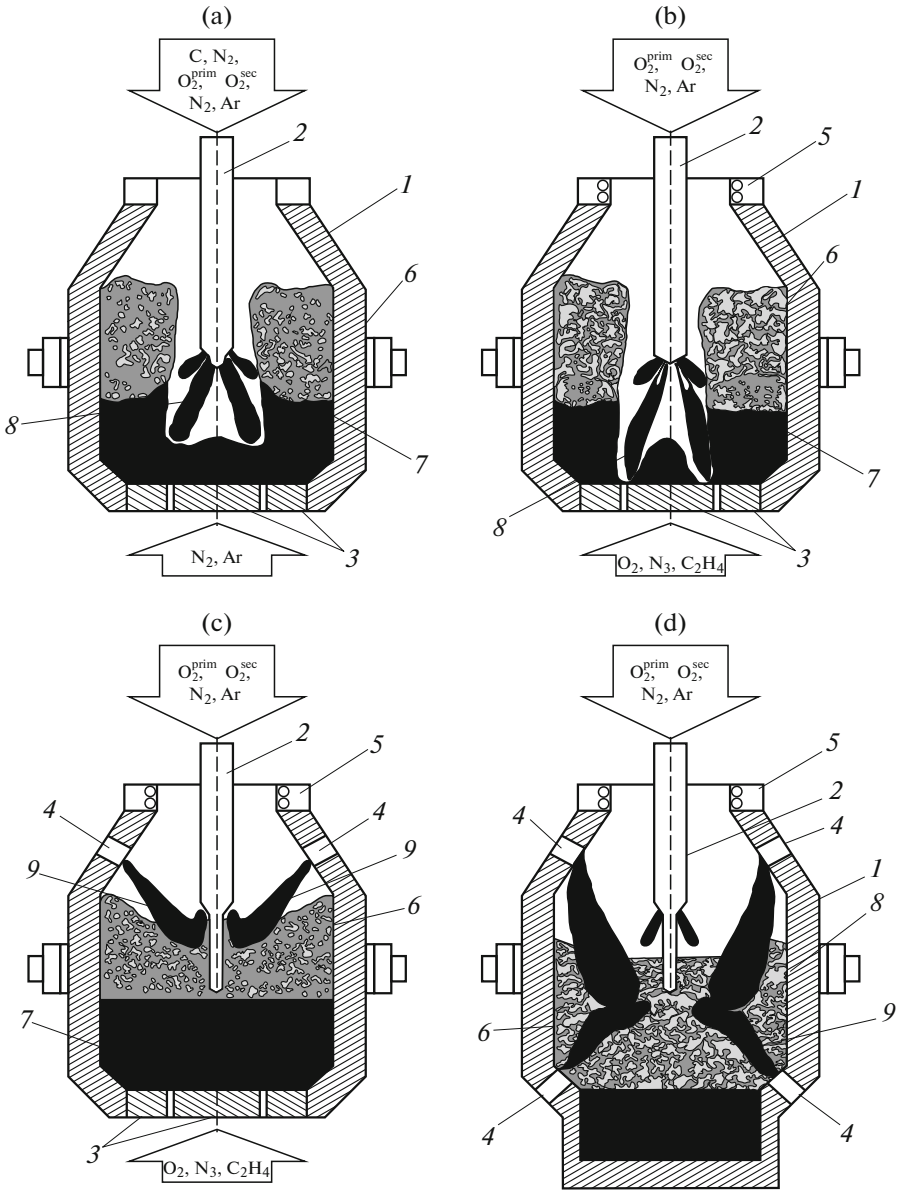

Fig. 1. Proposed converter systems and methods of complex bath injection: (*1*) converter; (*2*) multichannel top lance; (*3*) bottom lance for the supply of neutral mixing gases or natural gas; (*4*) lateral fuel–oxygen lance; (*5*) gas-heating system; (*6*) gas–slag–metal emulsion; (*7*) metal bath; (*8*) reaction zone of blast and exhaust gas; (*9*) fuel-combustion flame.

inexpensive manganese and chromium ores injected directly into the melt, rather than expensive ferroalloys [2, 3].

To increase the converter productivity and decrease the resource and energy consumption, we must reconfigure the system so as to permit preliminary heating of the scrap and batch heating and to intensify redox processes in the liquid bath and exhaust-gas combustion above the bath, without impairing the durability of the injection systems and the converter lining.

The new designs illustrated in Fig. 1 may be recommended for industrial adoption on the basis of laboratory and industrial experience with complex converter-bath injection [4–11] and the corresponding injection systems [12, 13]. Pulsed gas jets and fuel–oxygen combustion flames permit the solution of numerous technological problems [14–17]. It is important to assess the contribution of combustion flames to the redox processes in the converter bath.

The hydrodynamics in the reaction zones and in the liquid bath may obviously be greatly changed by fuel combustion in the converter's working space with jet formation or by means of submersible combustion flames. Note that in complex jets, over time, the gas temperature, volume, and composition will change. The volume of gas liberated will change not only on

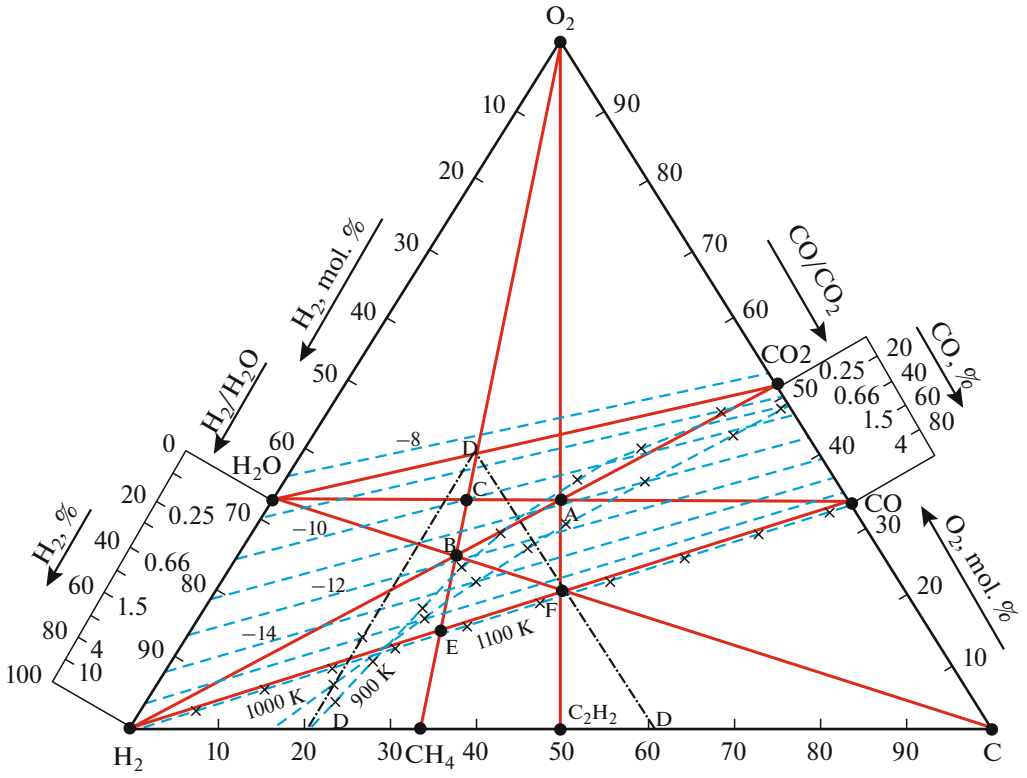

Fig. 2. Phase diagram of C–H_2–O_2 system: (– – × – –) isothermal boundary of the carbon deposition region at 1100, 1000, and 900 K; (---) isolines of $\log P_{O_2}$ at 1100 K.

account of the temperature rise, but also as a result of the chemical reactions, which may either increase or decrease the volume of the reacting phases and the system as a whole, depending on the circumstances.

The dynamics of gaseous-fuel combustion and the oxidation of elements in the converter bath on interaction with high-temperature combustion products may be analyzed by thermodynamic methods [19, 21].

Under certain assumptions, the interaction of the combustion flame and chemical elements in the converter bath may be calculated for equilibrium conditions. In practice, the equilibrium of individual reactions and the whole system in the smelting bath is virtually unattainable. However, equilibrium calculation reveals significant interrelationships in the behavior of elements in the converter bath and the balance in the reduction and oxidation of the impurities.

The interaction of system components in the combustion of gaseous hydrocarbon fuel may be established by analysis of the phase diagram of the C–H_2–O_2 system (Fig. 2). That permits quantitative estimation of the yield and composition of the reaction products at different stages of combustion 19].

We may identify three regions in Fig. 2. The H_2–H_2O–CO_2–CO region corresponds to four-component mixtures of the oxidation products ($CO + CO_2 + H_2O + H_2$) from the initial hydrocarbon mixtures $C_nH_m + H_2 + C$ and also small quantities of molecular oxygen ($P_{O_2} = 10^{-9}$–10^{-18} atm). Above this region, we note gas compositions consisting of the combustion products CO_2, H_2O, and O_2. Below, we note reducing gas mixtures consisting of CO and H_2, incompletely oxidized hydrocarbons, and solid carbon.

In the combustion of methane, with oxygen mixtures as the oxidant, the curve runs along the CH_4–O_2 line. In the combustion of other gases (for example, acetylene), the curve runs along the C_2H_2–O_2 line (or other mixtures of oxidant and hydrocarbon).

In production, the oxidation of natural gas, which consists mainly of methane, takes place in two stages. In the first, at 873–1473°C, the hydrocarbons are mainly converted by the reaction

$$2CH_4 + O_2 = 2CO + 4H_2. \quad (1)$$

In the second stage, carbon monoxide and hydrogen are oxidized by the reactions

$$2CO + O_2 = 2CO_2; \quad (2)$$

$$4H_2 + 2O_2 = 4H_2O. \quad (3)$$

Overall, the gas combustion in the combustion flames may be described by the reaction

$$(1/2)CH_4 + O_2 = (1/2)CO_2 + H_2O, \quad (4)$$

with

$$\Delta G^\circ_{(4)1650\,K} = -377907 \text{ J/mole } O_2;$$

$$\Delta G^\circ_{(4)2000\,K} = -355975 \text{ J/mole } O_2.$$

To obtain correct comparative estimates, all the reactions are converted to a single mole of oxygen O_2.

The quantity and composition of the reaction products depend primarily on the quantity of oxygen in the gas–oxygen mixture. In the absence of carbon and with insufficient oxygen—for example, if the gas–oxygen mixture corresponds to point E (27% C, 58% H_2, 14.5% O_2) at the intersection of the CH_4–O_2 and H_2–CO lines, when the O_2 content will be 16.8% of the methane content—the gas is converted in accordance with Eq. (1). The reaction products formed are H_2 and CO, in accordance with the length of the segments CO–E and H_2–E—that is, with an H_2/CO ratio (mol %) of 56.8 : 43.2. With increase in oxygen content in the gas–oxygen mixture to 25% (point B in Fig. 2), the oxidation of methane, in parallel with Eq. (1), also occurs by the reactions

$$CH_4 + O_2 = CO_2 + 2H_2; \quad (5)$$

$$(1/2)CH_4 + O_2 = (1/2)CO_2 + H_2O; \quad (6)$$

$$(1/3)CH_4 + O_2 = (2/3)H_2O + (1/3)CO. \quad (7)$$

The composition of the reaction products in the H_2–H_2O–CO_2–CO region and, correspondingly, the contributions of the reactions in Eqs. (1) and (4)–(7) are controlled by the equilibrium constant $K_{(E)}$ of the wet gas reaction

$$CO + H_2O = CO_2 + H_2,$$

$$K_{(E)} = \frac{P_{CO_2}}{P_{CO}}\frac{P_{H_2}}{P_{H_2O}}. \quad (8)$$

Here P_x denotes the equilibrium partial pressure of gas x, atm.

In addition, for gas mixtures with a high CO content, $\frac{P_{CO_2}}{P_{CO}}$ is controlled by the equilibrium constant of the reaction

$$2CO \Leftrightarrow CO_2 + C_{so}, \quad (9)$$

with the deposition of solid carbon (soot).

At low working temperatures (<1100 K), the region where carbon is deposited largely corresponds to the region occupied by multicomponent gas mixtures H_2–H_2O–CO_2–CO. Above 1100 K, the solid carbon may only be present in the H_2–C–CO region. With the possible increase in the H_2O concentration, we may observe the reaction

$$H_2O + C = H_2 + CO. \quad (10)$$

A universal characteristic of the state of the gas phase is the oxygen partial pressure, which may be determined from the equilibrium constants for Eqs. (2) and (3)

$$K_{(2)} = \frac{P^2_{CO_2}}{P_{O_2}P^2_{CO}}; \quad (11)$$

$$P_{O_2} = \frac{P^2_{CO_2}}{K_{(2)}P^2_{CO}}; \quad (12)$$

$$K_{(3)} = \frac{P^4_{H_2O}}{P^4_{H_2}P^2_{O_2}}; \quad (13)$$

$$P_{O_2} = \frac{P^2_{H_2O}}{P^2_{H_2}K^{1/2}_{(3)}}. \quad (14)$$

Thus, the thermodynamic parameters of the gas reactions may be characterized by $\Delta G^\circ = f(T) = -P\ln K_P$ or by the ratio of the equilibrium partial pressures $\frac{P_{CO}}{P_{CO_2}}$ or $\frac{P_{H_2}}{P_{H_2O}}$.

With complete combustion of methane to CO_2 and H_2O by Eq. (4), the initial gas–oxygen mixture must correspond to the point D, where the reaction products O_2 and CH_4 are formed in accordance with the length of the segments D–CH_4 and D–O_2, whose ratio is around 0.6. Specifically, the mixture must consist of 40.9 O_2 and 59.1% CH_4. (In composition, the mixture must correspond to 100% CH_4 and 69% O_2.) Thus, a gas phase with 40% CO_2 and 60% H_2O must be formed.

With less oxygen, the combustion of CO and H_2 (in the second stage of the process) will be incomplete. In other words, the gas phase will contain not only CO_2 and H_2O but also H_2 and CO. With more oxygen, the reaction products will also include free oxygen. In both cases, the total enthalpy of the process will be reduced: in the first case, on account of incompleteness of the exothermal oxidation of hydrogen and carbon monoxide; in the second, on account of heating of the excess oxygen. In the latter case, the excess oxygen may prove useful as thermal ballast in situations where the flame temperature must be reduced.

The characteristics of equilibrium oxidation of methane by oxygen, carbon dioxide CO_2, and water vapor at the temperatures in the converter bath (1800–2200 K) are presented in Fig. 3 and in Table 1. The notation employed is as follows: J, total enthalpy of the final products (referred to 1 mol of the initial

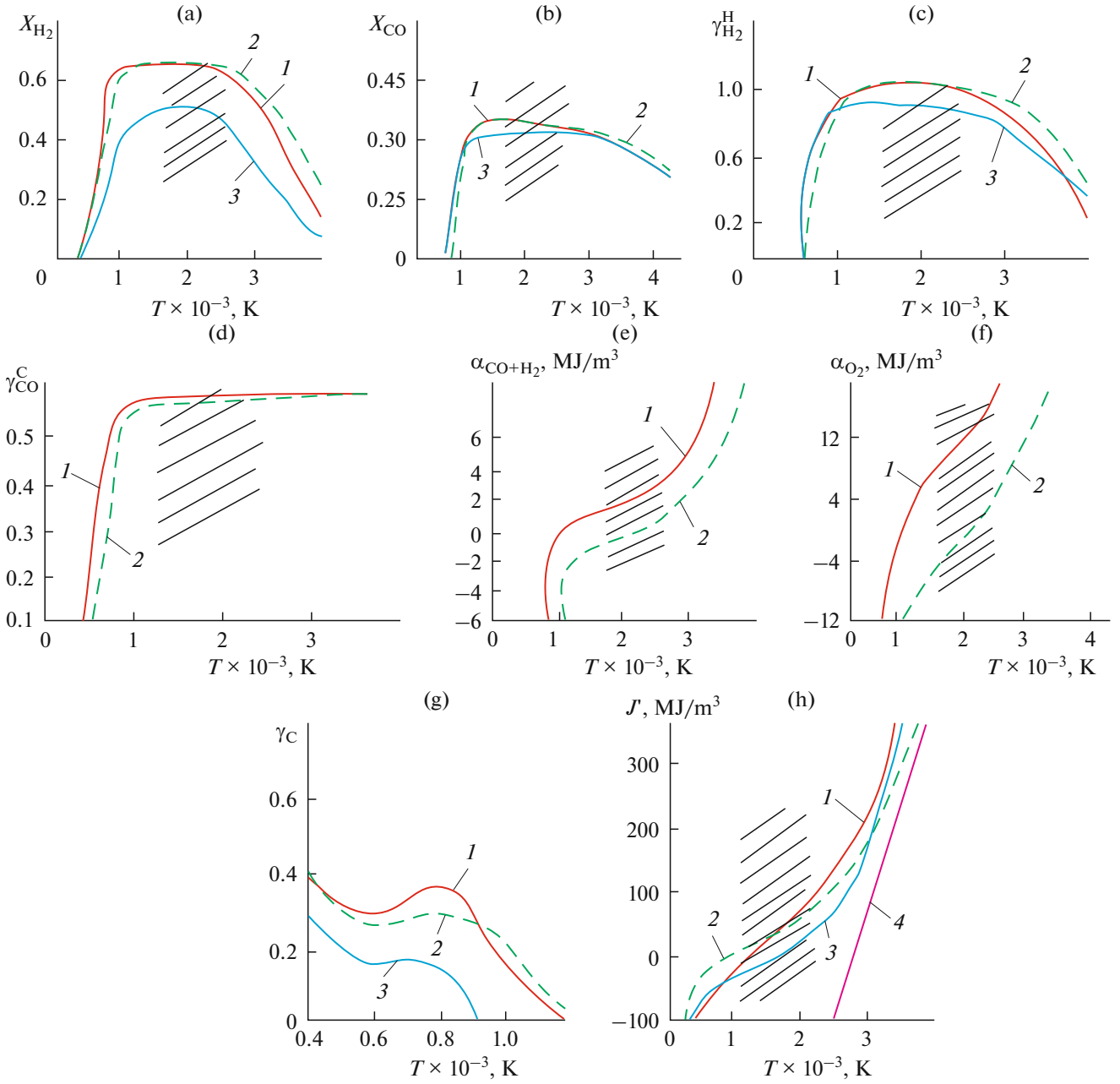

Fig. 3. Characteristics of equilibrium oxidation of methane by oxygen, when the initial reagent mixture is $CH_4 + 0.5O_2$ (*1*, *2*), $CH_4 + 0.65O_2$ (*3*), and $CH_4 + 2O_2$ (*4*); $P = 0.1$ MPa (*1*) and 0.3 MPa (*2*). The shading shows the temperature region of the flame in the converter's working space.

reagents), J/mol; γ^C_{CO}, degree of conversion of carbon in the initial reagents to CO; $\gamma^H_{H_2}$, degree of conversion of hydrogen in the initial reagents to molecular hydrogen; $\gamma^C_{CO_2}$ degree of conversion of carbon in the initial reagents to CO_2; $\gamma^H_{H_2O}$, degree of conversion of hydrogen in the initial reagents to H_2O; J', total enthalpy of the initial reagents, J/mol; α_{CO+H_2}, energy consumption referred to 1 m^3 of CO + H_2, kJ/m^3; $\alpha_{O_2+CO_2}$ (or α_{H_2O}), energy consumption referred to unit initial reagents O_2 + CO + H_2O, kJ/m^3 (or kJ/kg).

Note that, in the oxidation of methane by oxygen, all the reactions controlling the composition of the gas phase participate, including those in Eqs. (8) and (9). As a result, the levels of conversion of the carbon in methane to solid carbon, CO, and CO_2 and conversion of hydrogen to H_2O are low (5–20%) at low temperatures (1100–1200 K) and pressures $P = 0.1$ MPa.

Table 1. Characteristics of equilibrium oxidation of methane by oxygen, carbon dioxide, and water vapor ($P = 100$ kPa)

Characteristic	Value							
	$CH_4 + 0.5O_2$		$CH_4 + 2O_2$		$CH_4 + CO_2$		$CH_4 + H_2O$	
	1600 K	2200 K	1800 K	2200 K	1800 K	2200 K	1800 K	2200 K
Composition of products, mol; C : O = 1, H : C = 4,	C/O = 1	H/O = 4	C/O = 0.25	H/O = 1	C/O = 1	H/O = 2	C/O = 1	H/O = 6
H_2	0.6660	0.6628	0.0010	0.0070	0.4996	0.4970	0.7495	0.7457
CH_4	0.0002	–	–	–	0.0001	–	0.0002	–
H_2O	0.0001	–	0.6644	0.6467	0.0001	–	0.0001	–
CO	0.3332	0.3326	0.0019	0.0179	0.4999	0.4990	0.2499	0.2494
CO_2	–	–	0.3309	0.3106	–	–	–	–
O_2	–	–	0.0013	0.0109	–	–	–	–
$\sum H$	1.3340	1.3300	1.3310	1.3140	1.0000	0.9981	1.5000	1.4960
J	+10624	+25545	−232555	−191046	−7253	+7707	+19565	+34455
γ^C_{CO}	0.99860	0.99990	0.00565	0.05457	0.99970	0.99990	0.99930	0.99980
$\gamma^H_{H_2}$	0.998300	0.999100	0.001473	0.010650	0.999900	0.995800	0.999200	0.996600
$\gamma^C_{CO_2}$	0.000052	0.000100	0.994300	0.945400	0.000053	0.000010	0.000480	0.000009
$\gamma^H_{H_2O}$	0.002010	0.000052	0.998100	0.984300	0.000202	0.000053	0.000189	0.000048
J'	+21240	+51200	−222900	−19 390	−14500	+15440	+39120	+69070
α_{CO+H_2}	1586	2260	308400	29820	4905	5582	4409	5087
$\alpha_{O_2+CO_2}$	9511	13520	−13260	−11310	19610	22280	–	–

However, at the temperatures in the converter bath (1600–2600 K), practically 100% of the methane is converted to CO and H_2 (Figs. 3d and 3e) for a $CH_4 + 0.5O_2$ mixture. With increase in oxygen content to $CH_4 + 2O_2$, the degree of conversion is somewhat reduced; it also declines with increase in pressure to 0.3 MPa.

When C/O = 1, at 1200–2800 K, the final products consist mainly of molecular hydrogen H_2 and carbon monoxide CO (Fig. 3), as shown by thermodynamic analysis [20]. The CO and H_2 concentrations depend on the initial ratio of elements in the C–H–O system. When C/O = 1, the following formulas are of satisfactory accuracy [22, 23]

$$N_{CO} = \frac{2\left(\frac{C}{H}\right)}{1+2\left(\frac{C}{H}\right)}, \text{ mol};$$

$$N_{CO_2} = \frac{1}{1+2\left(\frac{C}{H}\right)}, \text{ mol}.$$

At low temperatures, the equilibrium products also contain CH_4, CO_2, H_2O, and C_{co} (condensed). The degree of conversion of the carbon in methane to carbon monoxide (γ^C_{CO}) and the degree of conversion of the hydrogen in methane to molecular hydrogen ($\gamma^H_{H_2}$) are about 100% when C/O = 1 and $T = 1500$–2500 K [22, 24].

With increase in pressure and increase in C/H, the region where condensed carbon exists (γ_C) expands. With increase in O/C, this region contracts.

In the oxidation of methane by carbon dioxide CO_2, the degree of conversion of the carbon in methane to carbon monoxide (γ^C_{CO}) at the temperatures in the converter bath declines to 0.6, while the degree of oxidation of hydrogen to H_2O rises to 0.95 (Figs. 3e and 3f). In oxidation by water vapor, by contrast, the degree of hydrogen conversion declines to 0.7.

CONCLUSIONS

In the converter production of steel, the use of the combustion flames is found to change the composi-

tion of the gas phase in the working space (above the bath): it contains H_2 and H_2O in addition to the traditional components O_2, CO, CO_2 obtained on oxygen injection. The presence of H_2 and H_2O changes the thermal conditions and oxidative properties of the gas phase.

In the combustion of gas–oxygen fuel, the optimal composition of the initial gas mixture (natural gas + oxygen) must correspond to the ratio 100% CH_4 + 69% O_2. The oxidation product is a gaseous phase consisting of 40% CO_2 + 60% H_2O.

The total enthalpy of combustion of the gas–oxygen fuel at converter temperatures, with an oxygen excess greater than 1.0 (up to 2.0), is about 200 kJ per mole of the initial reagents. In the oxidation of methane by carbon dioxide, the total enthalpy of combustion is between –7 and –14.5 kJ/mol of initial reagents at 1800 K. The process becomes endothermal at temperatures above 2000 K: $\Delta H_{2200} = 7.7–15.4$ kJ/mol. In the oxidation of gas by water vapor, $\Delta H_{1800–2200} =$ 19.5–70 kJ/mol.

Thus, flame temperatures above 1800 K may only be attained in the oxidation of methane by oxygen. The use of air, carbon dioxide, or water vapor as the oxidant does not yield the required thermal effect.

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