Abstract. The mixing and burning of natural gas with heated air in the reactor of carbon black obtaining have been analyzed. Commercial test results for the device of partially premixed components with single-flow burner have been considered. Construction peculiarities and operational conditions of single- and multi-flow burners have been examined.

Key words: burner, reactor, carbon black, natural gas, flame stability.

1. Introduction

The world output of carbon black exceeds 7 million tons per year [1]. Ukraine has high capacities of carbon black production in Kremenchuk and Stahanovsk. The development of new and advanced processes with simultaneous improvement of performance characteristics is of great importance.

At present carbon black is obtained by thermal conversion of hydrocarbon raw material in high-temperature flow of natural gas and air combustion products at majority of plants in the world, as well as at Ukrainian plants. The main parts of the reactor of carbon black obtaining are: a device for burning of natural gas with air, a mixing chamber for combustion products with hydrocarbon raw material, a reaction chamber, a chamber for reaction products cooling (quenching) till 1073 K using spray water. For the rational heat usage and reduction of natural gas specific consumption, air is preheated by gas-carbon mixture passing from the reactor after quenching zone.

The burning device consists of a burner, through which natural gas and air are supplied, and combustion chamber, where burning of natural gas with air takes place. The burner or its separate parts are made of steel. In order to achieve the stable and continuous operation of the reactor and maximum possible yield of carbon black the burning process must be organized in such a way that completeness of natural gas combustion, equal distribution of combustion products concentrations along cross-section before their mixing with hydrocarbon raw material, stable burning without flame-out or flashback and flame origin in the burner due to the self-ignition are ensured. Moreover, it is necessary to prevent the destruction of the burner metal parts occurring due to their burning or melting. A single burning device should guarantee a sufficiently high productivity (~1000–2000 nm³/h for natural gas and 10000–20000 nm³/h for air).

For the above-mentioned tasks solution it is advisable to choose the most acceptable burner type and to determine the device optimum geometrics as well as gas flow rates while burning of natural gas with heated air takes place. Different types of burners are widely used. They are classified by mixing method of gas flows, gas and air pressures before their feed to the burner, number of gas flows feeding to the burning zone, fuel gas calorific values etc. [2-4]. The choice of a burner type mainly depends upon gas flow parameters (pressure, temperature).

Usually at the plants of carbon black natural gas is supplied with overpressure not less than 3·10⁵ Pa and air is supplied with overpressure (0.4–0.5)·10⁵ Pa and preheated to 673–1073 K. Single- and multi-flow burners with preliminary mixing of the components and multi-flow diffusive burners without preliminary mixing are used [5-7]. Advantages and advisability of the mentioned burners used in the reactors of carbon black obtaining are open to discussion. Below we analyze conditions of natural gas with preheated air burning using burners of above-mentioned types.

2. Results and Discussion

2.1. Device with single-flow burner of preliminary mixed components for burning of natural gas with heated air

The principal scheme of the device is represented in Fig. 1.

The mixing and combustion chambers are lined with refractory and called as tunnels or tunnel burners [3, 8].
The device with single-flow burner of preliminary mixing is relatively simple in constructing and meets the mentioned demands under proper conditions of mixing and burning of natural gas with heated air. The burners with preliminary full or partial mixing may be used in the device. Chemically homogeneous mixture is obtained from the full mixing burner and kinetic burning takes place in the chamber. The burner with preliminary partial mixing ensures distribution of components small volumes along flow section and partial mixing at molecular level takes place simultaneously in the combustion chamber, i.e. diffusive-kinetic burning takes place. Under other equal conditions (temperature, pressure, flow rates, diameter of burner outlet) the mixing chamber length is several times less and combustion chamber is longer of the burners with partial mixing compared with those with full mixing. At high heating temperatures of air in the mixing chamber the temperature of natural gas with air mixture may be higher than self-ignition temperature. To prevent preignition of the mixture it is necessary that its residence time in the mixing chamber should be minimum possible. Therefore, it is advisable to use single-flow burner with partial preliminary mixing for natural gas combustion in the reactor of carbon black obtaining.

2.1.1. The choice of gas flow feed method to the burner

Different methods of gas flow feed (accompanying flows, counter-current flows, flow feed at an angle) to the burner are described in the literature [2, 3]. Since natural gas and air are fed to the reactor of carbon black obtaining under pressure, it is recommended to feed natural gas into the heated air coming to the mixing chamber throughout the holes of little diameters by jets. Such jet method of mixing allows to distribute small volumes of natural gas along cross-section of air flow for a very short period of time ($10^{-3}$ s). The main regularities of jet method of mixing are listed in [3, 4].

The penetration depth of jet fed to the continuous flow at the angle of 90° may be determined using Eq.(1):

$$h = k_s \frac{W_j}{W_f} \sqrt{\frac{\gamma_j}{\gamma_f}} d,$$

where $h$ – penetration depth of jet into the continuous flow; $W_j$ – jet linear velocity at the outlet; $W_f$ – flow linear velocity; $\gamma_j$ – density of gas fed by jet; $\gamma_f$ – density of gas flow; $d$ – diameter of a hole, throughout which the gas is fed by jet; $k_s$ – proportionality coefficient depending upon the distance between holes.

The jet develops in the continuous flow. The diameter of developed jet $D$ at the distance of penetration depth is $0.75h$.

Different variants of holes arrangement are used: in the walls of pipe, through which air is fed, in the walls of pipe placed along air flow axis etc. At Omsk carbon black plant in a single-flow burner natural gas is fed by a pipe through the holes situated in the walls of gas collector made in a form of beams which are joined to the pipe [7]. At Kremenchuk carbon black plant natural gas is fed into the air flow through the holes situated in the walls of gas collector made in the form of the ring with beams [9].
Such a design of feed chamber ensures equal distribution of natural gas small volumes along cross-section of the air flow in the mixing chamber of large diameter with corresponding large consumption of natural gas and air.

### 2.1.2. The choice of gas flow rate from the burner

While burning of preliminary prepared mixture (gas and air) in the described device (tunnel) stable burning (stable position of flame basis in the combustion chamber) is possible within definite limits of gas flow rates. If mentioned rate is less than a lower limit flashback takes place in the mixing chamber. If the rate is higher than the upper limit, flameout from the combustion zone takes place. The rate lower limit is called as flashback rate \( W_{fb} \) and the rate upper limit – flameout rate \( W_{su} \).

In accordance with theoretical and experimental results [10, 11] under turbulent conditions of gas flow in cylindrical pipe the flashback rate of methane-oxygen and methane-air mixtures may be determined from equation (2):

\[
W_{fb} = 1.78 \, Re^{0.1} \, U_n, \tag{2}
\]

where \( U_n \) – normal rate of flame spread; \( Re \) – Reynolds number of gas flow in the cylindrical pipe.

Reynolds number does not exceed value of 10⁶ in the mixing chamber. Pro tanto \( W_{fb} \) does not exceed \( U_n \) in more than 7 times. Under the conditions of carbon black obtaining the temperature of natural gas and air mixture which is fed to the combustion chamber does not exceed 1073 K. At mentioned temperature [8] the maximum value of \( U_n \) is 4 m/s (for the mixture with excess air coefficient \( \alpha = 1 \)). In accordance with Eq. (2) the flashback rate does not exceed 30 m/s.

The flameout rate depends upon physico-chemical properties of initial burning mixture (normal rate of flame spread, heat conductivity, heat capacity, flame temperature), gas dynamic characteristic of the flow in the combustion chamber, the chosen method of burning stabilization. In mentioned device the burning stability is achieved by circulation of high-temperature flows of combustion products to the gas flow of initial mixture at the burner outlet. Using such method of burning stabilization in tunnel burners while burning of cold methane-air mixture with \( a = 1.1–1.5 \) the flameout rate is 180–170 m/s, correspondingly [3]. It is mentioned in [3, 8] that under other equal conditions the flameout rate is proportional to the normal rate of flame spread to the degree nearly one. The normal rate of flame spread increases in 5 times with the increase of initial temperature of methane-air mixture from 293 to 673 K [8]. Hence, the flameout rate is more than 300 m/s while combustion of natural gas with air preheated to 673 K and higher temperatures. Thus, in described device a stable combustion is ensured within a wide range of flow rates from 30 to 300 m/s.

The gas flow rate at the burner outlet and correspondingly in the burner mixing chamber is admitted within a range of 60–200 m/s in the commercial reactors of carbon black obtaining to support the stable process without flashback and flameout. The higher gas flow rates are limited by permissible loss of pressure in the burner mixing chamber.

### 2.1.3. Determination of main geometric dimensions of the mixing chamber and combustion chamber

**Burner mixing chamber.** The diameter of a burner mixing chamber is determined by the assigned flow rate of combustible mixture and gas flow rate at the burner outlet. In order to achieve necessary productivity in the commercial reactor of carbon black obtaining at above-mentioned flow rates the diameter of a burner mixing chamber is within 0.2–0.3 m. The mixing chamber length may be comparable with the chamber diameter (0.7–1.0 of the diameter). At the jet feed of natural gas into air stream and mentioned length of mixing chamber the equal distribution of natural gas small volumes along cross-section of gas flow which leaves the burner outlet is achieved.

**Combustion chamber.** It is necessary to ensure flame stability and natural gas completeness of combustion in the combustion chamber. As mentioned above the stable burning (without flameout) is achieved by circulation of combustion products to the gas flow of initial mixture at the burner outlet. The circulation intensity and efficiency of burning stability depend upon ratio between diameters of combustion and mixing chambers. To prevent the flameout at high flow rate at the burner outlet it is necessary that the mentioned ratio would be within 2–4 [3, 8].

The combustion chamber length must be no less than turbulent flare length, at which completeness of natural gas combustion is achieved. A lot of experimental and theoretical investigations are devoted to the turbulent combustion of gas flows. The analysis and review of such works are presented in monographs [3, 8]. In general the dependence of turbulent flare length upon main factors may be described as follows:

\[
L_f = K_f \cdot d^n \cdot W^m \cdot U_n^p, \tag{3}
\]

where \( L_f \) – turbulent flare length, m; \( d \) – diameter of burner hole, m; \( W \) – rate of gas flow at the burner outlet, m/s; \( U_n \) – normal rate of flame spread, m/s; \( K_f \) – proportionality coefficient (its dimensionality \( m^{1-a} \cdot s^{-m} \cdot s^{-p} \) coordinates values of flare length in meters with other values).

The effect of temperature and combustion mixture composition on the flare length is evaluated using \( U_n \). Values
In accordance with experimental data presented in [12-14] for the kinetic burning of natural gas-air or natural gas-oxygen mixtures in developed turbulent flow of different diameters and rates in the tunnel the above-mentioned values are following: \( n = 0.8–0.9 \), \( m = 0.2–0.3 \), \( p = 0.45–0.5 \) and \( K_f = 1.1–1.2 \).

For the diffusion-kinetic burning the turbulent flare length also depends upon mentioned factors but values of exponents and \( K_f \) may differ from those of kinetic burning. Under similar conditions (burner diameter, temperature, gas and air flow rates) the length of turbulent diffusion-kinetic flare is larger than that of kinetic flare. The quantitative dependence of the length of turbulent diffusion-kinetic flare upon main factors is described in literature insufficiently. Therefore such length of the combustion chamber is needed that completeness of combustion would be guaranteed. The length is calculated mainly on the basis of experimental data. Based on the analysis of operating conditions of the reactor of carbon black obtaining using devices with single-flow burner of components partial mixing the general length of the combustion chamber is 6-8 times larger than the diameter of a burner hole.

It should be noted that turbulent flare length is proportional to the diameter of a burner hole to the degree nearly one and insignificantly depends upon gas flow linear velocity for the kinetic burning as well as for the diffusion-kinetic one. Insufficient effect of linear velocity increase on the change of turbulent flare length is caused by the fact that increase of linear velocity increases the turbulent diffusion coefficient, turbulent rate of flame spread and decreases time of burning completion in the turbulent flow. Therefore devices with assigned geometrical dimensions of the mixing and combustion chambers, the completeness of combustion changes slightly with the change of linear velocity and combustion mixture flow rate within wide interval (in 1.5–2 times).

2.1.4. Analysis of self-ignition conditions of heated mixture of natural gas and air in the burner mixing chamber

Natural gas containing mixture of ethane and propane (10–12 vol %) together with some amount of C_2–C_3 hydrocarbons as well as methane is used for production of carbon black in Ukraine. A heated mixture of gaseous alkanes with air is formed in the burner mixing chamber. The excess air coefficient \( \alpha \) in the mixture may be within 1.0–1.5.

Self-ignition of the gas-air mixture may occur in the case, when mixture residence time in the mixing chamber is equal or greater than induction period for assigned temperature, pressure and conditions of mass- and heat transfer. It is necessary to take into account that ideal displacement regime of gas flow movement is not fully reached and residence time of individual elementary volumes of combustible mixture, in which self-ignition is possible, may be greater than average residence time of the gas mixture in the chamber. To prevent self-ignition of combustible mixture the average residence time of gas flow in the mixing chamber would be nearly 5 times less than self-ignition induction period for assigned temperature.

As it was shown above, the gas flow rate in the mixing chamber of the industrial burner is within 60–200 m/s and the mixing chamber length is 0.2–0.3 m. The maximum average residence time of combustible mixture in the burner mixing chamber is 0.005 s. To prevent premature flame origin in the mixing zone self-ignition induction period would be more than 0.025 s.

Accordingly to literature data [15, 16] self-ignition induction period of mixtures of C_1–C_4 alkanes with air or oxygen at 853–873 K and atmospheric pressure is nearly one second. It is obvious that self-ignition of natural gas-air mixture in the burner mixing chamber is impossible at mentioned temperatures. At higher temperatures (> 973 K) experimental values of self-ignition induction period of gaseous alkanes mixtures with air or oxygen are absent in literature. The self-ignition induction period decreases with the temperature increase and may be calculated from Eq. (4):

\[
\tau = Ae^{\frac{E}{RT}},
\]

where \( \tau \) – self-ignition induction period of the mixture with assigned composition at temperature \( T \), \( s \); \( T \) – mixture temperature, K; \( E \) – effective activation energy, J/mol; \( R \) – absolute gas constant = 8.314 J/mol·K; \( A \) – constant, s.

In [15] there are experimental values of self-ignition induction period of mixtures of C_1–C_4 with oxygen determined within 673–873 K at atmospheric pressure. Effective activation energies are calculated on the basis of obtained data. At constant temperature self-ignition induction periods change slightly with the change of excess air coefficient \( \alpha \) within 1.0–1.5. The effective activation energy \( E \) is ∼250 kJ/mol for the methane-oxygen mixtures and \( E = 160 \) kJ/mol for the mixtures containing ethane and propane (to 10 vol % calculated for methane) in addition to methane. Under other similar conditions (temperature, pressure and excess air coefficient \( \alpha \)) self-ignition induction periods of the mixtures of combustible gas with air or oxygen slightly differ from each other [16].

Results obtained in [15] with valid approximation allow to determine the maximum temperature of natural gas-air mixture at which self-ignition in the burner mixing chamber does not occur. For the mixtures of natural gas with oxygen or air which contain the mentioned amount
of ethane and propane in addition to methane the calculated values of self-ignition induction period are: 0.06 s – at 973 K, 0.03 s – at 993 K and 0.016 s – at 1013 K. To prevent self-ignition the temperature of natural gas-air mixture should be less than 993 K.

At the carbon black plants only air is mainly heated. Natural gas is fed into a reactor with the temperatures of 293–303 K. In order to achieve mixture temperature of 993 K the air should be heated to 1073 K. Thus, in accordance with adjusted results self-ignition of natural gas-air mixture will not occur in the burner mixing chamber at air heating to 1073 K.

2.1.5. Test of device with single-flow burner of components partial premixing for the combustion of natural gas with heated air in the commercial reactor of highly active carbon black obtaining

Reactors with mentioned device have been widely used at Kremenchuk carbon black plant for two years. Jet mixing is used in the reactor burner. Natural gas is fed into air continuous flow through the holes with different diameters situated in the tube walls of metal gas collector made as a ring with beams. In accordance with above-mentioned dependences of the jet penetration depth into a continuous flow and diameter of developed jet upon main factors the holes optimum dimension and distance between them were calculated. Equal distribution of natural gas small volumes along cross-section of air flow is achieved at these values.

Scheme of the reactor with described device is presented in Fig. 2.

The ratio between geometrical dimensions of the burner mixing chamber and combustion chamber agrees with the above-mentioned dependencies. At the same time it is necessary to pay attention to some constructive peculiarities of the combustion chamber. In the reactors of highly active carbon black obtaining a diameter of the mixing chamber of combustion products with hydrocarbon raw material must be considerably less than that of the combustion chamber after the burner. In connection with this fact the outlet part of combustion chamber looks like truncated cone finished by the cylinder. The cylinder diameter is the same as that of the mixing chamber of hydrocarbon raw material with combustion products. The diameter of inlet part is less than the diameter of central part of combustion chamber, it may additionally turbulize the flare in the burning zone.

The device for natural gas combustion was tested under reactor hot bank regime. Under such regime only burning process was supported, the hydrocarbon raw material was not fed to the reactor. Before the reactor outlet combustion products were cooled in the quenching zone by spray water. The composition of natural gas fed to the reactor was following (vol %): CH$_4$ – 86.3, C$_2$H$_6$ – 7.3, C$_3$H$_8$ – 3.0, C$_4$H$_{10}$ – 0.9, C$_5$H$_{12}$ – 0.1, N$_2$ – 1.3 and CO$_2$ – 1.1. Operating performance: natural gas consumption –

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Fig_2.png}
\caption{Scheme of the reactor of carbon black obtaining with device for the burning of natural gas with air: metal gas collector (1), mixing chamber of natural gas with air (2), combustion chamber (3), mixing chamber of combustion gases with raw material (4), reaction chamber (5), quenching chamber (6). Flows: natural gas (I), air (II), raw material (III), water (IV), mixture of reaction gases with carbon black (V). Indexes: $d$ – diameter of the burner mixing chamber (hole), $D_1$ – diameter of the inlet cylindrical part of combustion chamber, $D_2$ – diameter of the central cylindrical part of combustion chamber, $D_3$ – diameter of the outlet cylindrical part of combustion chamber, $L_{mc}$ – length of the burner mixing chamber, $L_{cc}$ – combustion chamber length.}
\end{figure}
980 nm$^3$/g, air consumption – 12400 nm$^3$/g, water consumption for the quenching of combustion products – 4200 l/g, natural gas temperature – 293 K, air temperature – 723 K, temperature of lining wall in narrowed part of combustion chamber (measured by pyrometer) – 2053 K, temperature of combustion products after quenching – 1103 K, reactor pressure – 0.027 MPa.

At mentioned consumptions of gas flows and composition of natural gas the excess air coefficient is 1.2. Calculated adiabatic temperature of combustion products is 2353 K. The gas flow rate at the burner outlet is more than 100 m/s.

To determine the completeness of natural gas combustion and equal distribution of components concentration along the cross-section of combustion products in the end of burning zone samples of combustion products were taken by special sampling device cooled by water. Samples were taken through the holes of raw jets in the narrowed part of combustion chamber at different distances from chamber walls. The composition of combustion products (residue gas) was determined by chromatography and presented in Table. The analysis sensitivity was 0.01 vol %.

One can see from the Table that methane is absent in combustion products and amount of incomplete combustion products (CO and H$_2$) is miserable. 0.2 vol % of H$_2$ presented in Sample 3, taken from the middle of the chamber, is perhaps connected with water vapors dissociation at high temperatures of gas flow in this place. A difference of CO$_2$ and O$_2$ concentrations in various samples does not exceed 1.5 vol %. The excess air coefficient $\alpha$ is within limits 1.14–1.21. At chosen geometrical dimensions of device for the combustion and assigned consumptions of gas flows the complete combustion of natural gas and equal distribution of components concentration along cross-section of combustion products flow before their mixing with hydrocarbon raw material are ensured.

The experiment of reactor long-term commercial operation shows that stable combustion without flashback, flameout and flame vibration (buzzing) is achieved using above-mentioned device for the combustion of natural gas with heated air. The burning of metal gas collector is prevented because direct contact between its surface and high-temperature flow of combustion products is absent. Described device ensures long-term, persistent and stable operation of the reactor of carbon black obtaining.

### 2.2. Multi-flow burners with partial premixing of combustible gas with air

Similarly to the single-flow burners it is possible to ensure completeness of combustible gas combustion and stable combustion for the multi-flow burners by choice of the optimum geometrical dimensions, consumptions and gas flow rates. The increase of flow amount will allow to achieve high productivity of the device at relatively short combustion chamber.

Nevertheless it is a problem of multi-flow burners to achieve equal distribution of natural gas and air consumption by separate flows. Moreover, the completeness of combustion may be achieved only at larger excess air coefficient compared with that of a single-flow burner. Multi-flow burners are more complicated in manufacture. So, it is more advisable to use single-flow burners with partial premixing of the components.

### 2.3. Device with the burner without components partial premixing for the combustion of natural gas with air

The mixing and combustion of combustible gas with air take place simultaneously in the combustion chamber using the burners of such type, i.e. diffusion burning takes place. The flame spread rate in turbulent gas flow is defined by turbulent diffusion rate. In accordance with literature data [3, 8] the length of diffusion turbulent flare is proportional to the diameter of single gas flow fed to the combustion chamber in degree more than one (1.2–1.5). It is also proportional to the linear velocity of gas flow in

### Composition of combustion products (residue gas) in the samples taken at the outlet of combustion chamber

<table>
<thead>
<tr>
<th>Sample number</th>
<th>CO$_2$</th>
<th>O$_2$</th>
<th>CO</th>
<th>H$_2$</th>
<th>CH$_4$</th>
<th>N$_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>9.8</td>
<td>4.1</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>86.0</td>
</tr>
<tr>
<td>2</td>
<td>10.2</td>
<td>3.4</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>86.4</td>
</tr>
<tr>
<td>3</td>
<td>10.6</td>
<td>2.7</td>
<td>0</td>
<td>0.2</td>
<td>0</td>
<td>86.5</td>
</tr>
<tr>
<td>4</td>
<td>10.1</td>
<td>3.6</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>86.3</td>
</tr>
<tr>
<td>5</td>
<td>9.9</td>
<td>4.0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>86.1</td>
</tr>
</tbody>
</table>

Note: Samples 1 and 5 were taken near chamber walls on the opposite sides. Sample 3 was taken from the middle of the chamber. Samples 2 and 4 were taken at the distance of 1/4 $D_3$ from chamber walls on the opposite sides.
3. Conclusions

On the basis of carried out investigations optimum geometrical dimensions of the device with single-flow burner of partial premixing of initial components have been determined. Gas flow rates have also been determined, at which the flame stability and completeness of combustion of natural gas with heated air are ensured in the reactor of carbon black obtaining. It is advisable to use single-flow burners with components partial premixing and the air heating to 1073 K. At higher temperatures multi-flow burners without premixing are used.

References
