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EFFECT OF MOISTURE ON NITROGEN DIOXIDE FORMATION IN LAMINAR FLAME OF NATURAL GAS

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Highlights

▶ Water vapor is the third very influential participant along with hydrocarbon fuel and oxidizer in the combustion process.

▶ Concept of the "excess moisture ratio" in combustion process is introduced.

▶ Effect of moisture in combustion process is due to HO_2^- and OH^- radicals.

▶ HO_2^- radical plays a crucial role in the oxidation of NO to NO_2 in natural gas flame.

▶ Injection of moisture into the combustion process has a significant effect on the formation of "fast" ("kinetic") nitrogen oxides.

Abstract. The paper contains the results of experimental studies of the effect of moisture on nitrogen dioxide formation and on oxidation of NO to NO_2 in laminar premixed flame of natural gas.

The water vapor is shown to be the third very influential participant, along with fuel and oxidizer, in the combustion process. Injection of moisture into the combustion zone has an effect due to the insertion of additional quantities of HO_2^- and OH^- radicals into the process, which contributes to the intensification of the oxidation of NO to NO_2. Introduction of the concept of the "excess moisture ratio" in the combustion process is proposed. The studies were executed at the laboratory installation in conditions of formation of the V-shaped laminar flame of natural gas behind a transverse cylindrical steel stabilizer, with determining the concentrations of flue gas components.

Keywords: burning process, nitrogen dioxide, moisture, peroxide radical, air pollution.

Introduction

The valid European regulations (European Parliament & Council of the European Union, 2010, 2015) impose strict limits on nitrogen oxide emissions for fuel-burning equipment. One of the most common methods for reducing emissions of nitrogen oxides is ballasting the combustion zone to reduce the maximum local temperatures and, accordingly, to reduce formation of the nitrogen monoxide.

As a ballast, the recirculation gases or inert gases are most commonly used, and recently more often the water vapor is; the latter is even more effective ballast. However, it is necessary to take into account that moisture takes part in the combustion process, causing it to change.

In particular, injection of moisture increases the proportion of nitrogen dioxide NO_2 in the total content of nitrogen oxides NO_x in flue gases. The NO_2 is much more toxic than monoxide NO (the maximum permissible concentrations are 0.085 mg/m^3 and 0.4 mg/m^3, respectively), but it is well soluble in water, in contrast to poorly soluble NO.

Decreasing of the equivalent diameter of the boiler combustion chamber leads to the increase in the specific content of NO_2 in the combustion products. The typical share of NO_2 is no more than 5% in the flue gases of large boilers of thermal power plants (usually 15…20 mg/m^3 of NO_2 and 480…500 mg/m^3 of NO) and is maximal (up to 25–30% of the total NO_x content, usually 60…90 mg/m^3 of NO_2 and 160…200 mg/m^3 of NO) in small household boilers (Sigal, 1994; Sigal et al., 2020). Regardless of the size of a boiler, the nitrogen dioxide content in the combustion products increases with reducing of load, increasing of the excess air ratio and of the degree of shielding of the furnace (Sigal, 1988).

Most of the medium size boilers operate in hot-water mode and are not equipped with economizers for flue gases heat recovery. The temperature of gaseous emissions from these boilers reaches 450 K and more. In order to improve their energy efficiency, these boilers are increasingly equipped with heat utilizers. Installation of the condensing heat utilizers is the most effective way as it enables to solve several important tasks:

– the thermal pollution of the atmosphere is reduced;
– not only the heat losses with flue gases (q_2), but also the internal heat of water vapor condensation are returned to the energy process, which increases the fuel utilization factor (FUF);
– the condensing product is distilled water, which can subsequently be used as feed water for boilers;
– the greenhouse gas emissions of the water vapor and partially of CO_2 is reduced.

At the same time, the authors found that application of a contact condensing heat utilizer contributes to solving the one more problem, namely reduction of the nitrogen oxide emissions into the atmosphere (Aronov & Sigal, 1988; Sigal & Paderno, 1992).

The combined condensing heat recovery technology, developed by the author (Primak & Sigal, 1992), combines in a single apparatus both the absorption cleaning unit which operates by dissolving NO and NO_2 in water, and the contact heat recovery unit, as well as the contact air heater as component of this apparatus or as the separate one (Figure 1).
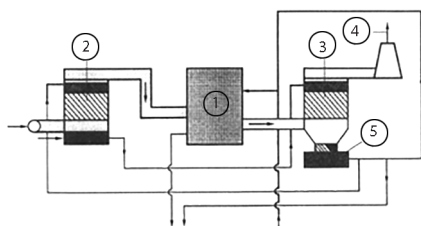

Figure 1. Scheme of the combined condensing heat recovery unit: 1 – boiler; 2 – contact air heater; 3 – contact heat recovery unit; 4 – chimney; 5 – carbon removing section

In the contact heat exchanger, the hot flue gases pass through a layer of packing holding the water film on its surface. The gases give their heat to water and are released into the atmosphere with a temperature close to 330K. The blast air passes through the contact air heater, gets heated up in it and is saturated with moisture, that causes its increased and constant humidity.

The experimental study of operational characteristics of two types of packing – Raschig ceramic rings and the developed construction with using the ribbon of amorphous metal alloy (Sigal & Paderno, 2020) has shown that the last one surpasses the ceramic one by the basic operational heat engineering parameters and contributes to further oxidation of the NO to water-soluble NO_2, which enables to remove more of the latter by absorption with water and, as a result, to reduce the emissions of nitrogen

oxides to the environment after contact apparatus by 35% more efficiently than with using the ceramic packing.

Thus, the combined system enables both reducing of the formation of NO in the process of fuel combustion with a simultaneous increase of NO_2 portion in combustion products by affecting the regime factors, and subsequent neutralization of nitrogen dioxide via absorption, and at the same time provides for the heat recovery that enables to improve the economy of removing of nitrogen oxides from the flue gases with partial payback of environmental protection activities.

It is known that the best solubility of NO_2 is in the cold water, whereas for heat utilization the higher temperature is better. This dilemma is treated either in two separate devices or in the single one by solving the optimization task.

The proposed approach is becoming increasingly relevant, since the thermal modernization of buildings leads to decrease in boiler loads, and decrease in load in turn increases the portion of NO_2.
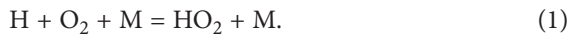
This work is an attempt to quantify the effect of moisture on formation of the nitrogen oxides in the combustion process. Of particular interest is the determination of the effect of moisture content on the process of additional oxidation of the nitrogen oxide NO to the nitrogen dioxide NO_2, since an increase in the portion of the well soluble in water nitrogen dioxide in flue gases, despite its 5 times greater toxicity than of insoluble NO, with its subsequent neutralization through solution in a contact apparatus will reduce the total toxicity of gas emissions, that allows to meet the requirements of regulatory documents.

Analysis of publications

The presence of moisture in combustion zone modifies the temperature field in the combustion space, causing changes of the conditions for nitrogen dioxide formation and dissociation in the process. A number of publications are devoted to the investigation, both theoretical and experimental, of the effect of moisture on the nitrogen oxides formation in the natural gas flame (Lukoshyavichus et al., 1986; Soroka et al., 2018; Roslyakov, 1988; Göke & Paschereit, 2013; Glarborg et al., 2018; Lamoureux et al., 2016; Shahpouri & Houshfar, 2019; Soroka & Zgurskyi, 2019; Navrodska et al., 2019). However, the contradictory findings on the efficiency of water injecting into the combustion zone to suppress the nitrogen oxides formation indicate a significant diversity of conditions of water injecting on the one hand, and on the other, an insufficient knowledge of the effect of moisture on the nitrogen oxides formation and decomposition.
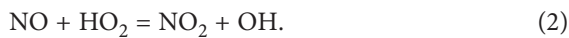
Research carried out by the author (Sigal, 2004; Sigal & Bykorez, 2007; Sigal & Dolinsky, 2007; Sigal et al., 2019) have demonstrated that the nitrogen dioxide formation occurs in the pre-flame and post-flame zones at temperatures up to 1000 K.

The main oxidizing agent of NO into NO_2 is the peroxide radical HO_2^- under strong flame cooling. It is formed by the following reaction:

$$H + O_2 + M = HO_2 + M. \tag{1}$$

The rate constant of this reaction is $3.3 \times 10^{-29} \times T^{-1.24}$ (Kondraťev, 1970).

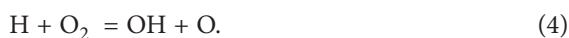Then HO_2 additionally oxidizes NO by the reaction:

$$NO + HO_2 = NO_2 + OH. \tag{2}$$

The oxidation process is limited only by the amount of HO_2^-, since concentration of NO is substantially higher than of HO_2^- radicals.

Significant quantities of peroxide radical are formed in hydrocarbon flames in the zone of disappearance of atomic hydrogen and near it. The entire amount of peroxide radical, taken out of the combustion zone as a result of the diffusion processes into the pre-flame and post-flame zones, reacts with the NO_2 formation.

These reactions are significant only in the low-temperature zone of a flame. In the temperature zone above 1000 K, the further oxidation of NO to NO_2 is possible by the reaction:

$$NO + OH = NO_2 + H \tag{3}$$

with using hydroxyl OH formed by the reaction:

$$H + O_2 = OH + O. \tag{4}$$

Rate constant is $2.56 \times 10^{-10} \times \exp^{-8365/T}$ (Kondraťev, 1970).

The results of experimental work have demonstrated that moisture injected with natural gas and blast air into the combustion process can be divided into:

– moisture that is involved in the chemical process of fuel oxidation, i.e. the one that decomposes during high-temperature dissociation into oxygen and hydrogen, followed by formation of the hydroxyl OH^- and the peroxide radical HO_2^-:

$$H_2O \leftrightarrow OH + H; \tag{5}$$

$$H + O_2 \rightarrow HO_2, \tag{6}$$

– moisture that is not involved in chemical processes of combustion, i.e. the excess moisture, the one that plays only the role of thermodynamic ballast and removes heat from the combustion zone (Soroka & Zgurskyi, 2019).

The ratio of all moisture in the process of burning W_{gen} to the one that is involved in the chemical processes of burning W_{chem} is proposed to be introduced as the "*excess moisture ratio*":

$$\beta = W_{gen}/W_{chem}. \tag{7}$$

Injection of the additional moisture increases the amount of the peroxide radical HO_2^- in the post-flame zone and, accordingly, due to this leads to the additional oxidation of NO to NO_2.

Research objectives:

– Experimental study of the effect of moisture on the nitrogen oxides formation and the further oxidation of NO to NO_2 in a laminar flame of natural gas while burning a three-component mixture (fuel, oxidizer, water).

– Experimental study of the effect of the peroxide radical HO_2^- on the additional oxidation of NO to NO_2 in a laminar flame of natural gas.

1. Research methods

The researches were carried out with a laminar flame, because it allows to illustrate physical and chemical patterns most clearly. The studies were executed at the laboratory installation, with formation of the "V-shaped" laminar premixed flame of natural gas, split by the transverse cylindrical steel stabilizer (Figure 2).

The width of the flame front did not exceed 2.5 mm. An area of 0.5 mm ahead of the flame front was also examined. The absence of an abrupt local temperature change made it possible to minimize the measurement error related to the inertia of the thermocouple.

The measurements were carried out on the V-shaped natural gas flame by parallel moving the probe and the thermocouple mounted in the clamps at an angle of attack to the flame front 90° with a step of 0.1 mm. The measurements were performed from the side of the pre-flame zone, then inside the flame front and in the post-flame zone. In each experiment, 6 passes were made, and up to 30 measurements were executed along 3 mm working range, with 6 repeats of each point respectively. Data from 6 measurements were averaged.

1.1. Description of experimental installation

The experimental installation consisted of the gas pipe equipped with air and gaseous fuel supply systems, the gas burner unit, the system for feeding droplet moisture into the flame front with capillary nozzle, the system for precise movement of the gas sampler, the thermocouples and the system for monitoring of the parameters of the combustion process (Figure 2).
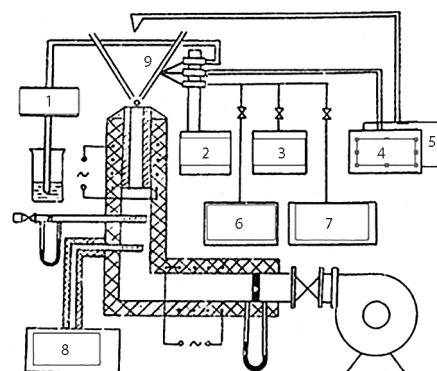

Figure 2. Layout of the experimental installation:
1 – peristaltic pump; 2 – precision positioning unit;
3 – sample preparation device; 4 – recorder; 5 – voltmeter;
6 – portable chemical gas analyzer; 7 – gas analyzer;
8 – flowmeter; 9 – flame

The gas pipe is embodied as the heat-insulated cylindrical tube made of refractory material with 16.0 mm

diameter and 4.0 mm wall thickness. Blasting air fan is attached to the outer end of the horizontal section of the tube through the hose long enough for stabilization of the flow (about 3 m), with condensate remover; the last one is particularly necessary in cases when the water vapor was fed specially for increasing humidity of the blast air to provide the equal humidity in all experiments. In the horizontal section there is a diaphragm with a U-like micromanometer for measuring the air flow. Gas burner unit is placed at the end of the vertical section of the gas tube. A nozzle for supplying the mixture of gaseous fuel and air, with system for heating the mixture to a predetermined temperature, is installed in the vertical section.

The gas is supplied through a copper tube of 10 mm in diameter, in which a measuring diaphragm with a U-like micromanometer is installed to measure the gas flow. To regulate the flow of air and gas, precision needle valves are installed.

The gas burner unit ensures complete premixing of gas with air.

Examination of the normal combustion front in the experiments was carried out in a stationary "V-shaped"' split flame, which is formed when the mixture is continuously fed into the burner behind a transverse cylindrical steel stabilizer of 3.0 mm in diameter (Figure 3). In preliminary experiments it was discovered that within the diameter of stabilizers ranging from 1 to 5 mm the elementary structure of the flame front does not depend on the diameter of the stabilizer.
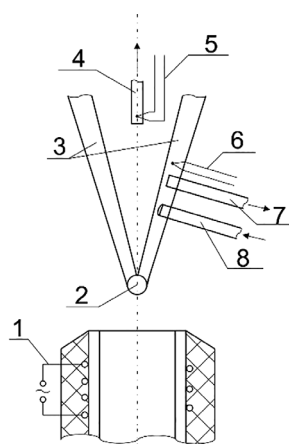

Figure 3. Scheme of the V-shaped flame formation and probes disposition: 1 – heater; 2 – transverse cylindrical stabilizer; 3 – V-shaped split flame; 4 – fixed gas sampler; 5 – fixed thermocouple; 6 – mobile thermocouple; 7 – mobile gas sampler; 8 – moisture-feeding capillary

In order to minimize the perturbation of the flame front, the gas sampler needle was sharpened to outer diameter 0.08 mm, and thermocouple junction – to outer diameter 0.03 mm. The distance between the moisture-feeding capillary (8) and the flame stabilizer (2) was 8 mm, the mobile thermocouple (6) was placed closely alongside with the mobile gas sampler (7) at the distance about 3 mm from the moisture-feeding capillary (8), the positions

of the fixed gas sampler (4) and the fixed thermocouple (5) were nearby along the axis of the gas supply tube at 20 mm above the flame stabilizer.

When the excess air ratio α changed from 1.03 to 1.8, the width of the flame front did not exceed 2.5 mm. The flow rate of the mixture was 1500 mm/s.

In order to ensure supply of droplet moisture into the flame front, the peristaltic pumps were used, which were connected to the source of moisture and to the capillary with silicone rubber hoses. Moisture-feeding capillaries with an outer diameter of 0.8 mm were made of stainless steel. Peristaltic pumps were equipped with a flow rate regulator, which allowed to stabilize moisture flow at a given level, to ensure compensation for the moisture evaporated in the flame, thus the volume and surface of evaporation of the droplet injected into the laminar flame front were kept unchanged.

1.2. Method for determining the concentration of flue gas components

The sampling system consisted of the following main elements: the gas sampler, the line through which the sample entered into the analyzing device, the refrigerator used to remove moisture from the sampled gas, and the filters that catch dust and soot particles.

The gas sampler was made of stainless steel, which minimized the catalytic effects leading to the oxidation of NO to NO_2 on the walls of the gas sampler. Both cooled and uncooled samplers were used. The nozzle of the sampler had a streamlined shape and, therefore, insignificantly distorted the flow field in the measurement area. The length of the sampler nozzle was chosen in such a manner that the disturbances arising during the flow around the holder would not affect the flow field at the measurement point.

Diameter of the sampler nozzle was about 1 mm, which ensured the high rate of temperature falling of the gas sample due to contact with the cold walls of the sampler.

In order to estimate the rate of gas temperature falling, the flow regime, which was formed in the sampler, was estimated. Thus, at the flow rate of the sampled gas through the analyzer about 15 cm^3/s, the velocity at the inlet to the sampler was about 7 m/s.

In experimental studies, the focus was placed on measuring the maximum local temperature and the corresponding concentrations of nitrogen oxides in the "trace" of vaporization of droplet of the water or of the hydrogen peroxide solution.

2. Results and interpretation

2.1. Dependence of the distribution of temperature fields and nitrogen oxides concentrations on humidity across the width of the flame front

The researches elicited data on the distribution of temperature fields and nitrogen oxides concentrations across

the width of the flame front in dependence of the amount of additional moisture injected (H_2O droplets):

 a) without injecting a drop, at the constant $\alpha = 1.1$;
 b) without injecting a drop, under α changing from 1.03 to 1.22;
 c) with injecting drop of various diameter ranging from 1.0 to 4.0 mm, at the constant $\alpha = 1.1$;
 d) with injecting drop of the constant diameter ($d = 2.0$ mm), under changing α from 1.03 to 1.22;
 e) with injecting two drops into the flame front ($d = 1.0$ and 2.0 mm), at the constant $\alpha = 1.1$.

When α changes from 1.03 to 1.22, the maximum temperature of the flame front decreases from 1573 to 1373 K without injecting a drop and from 1323 to 1123 K when drop is injected (Figure 4).
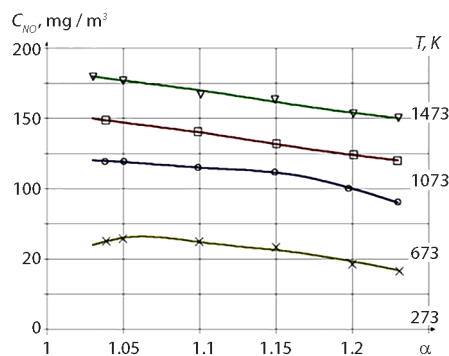

Figure 4. NO concentration and temperature in the front of the laminar flame of natural gas: ∇ – T_0 – temperature without injecting moisture; $\bigcirc$ – NO_0 – NO concentration without injecting moisture; $\square$ – T_1 – temperature with injection of one H_2O droplet; x – NO_1 – NO concentration with injection of one H_2O droplet

Decreasing of temperature with changing α both with and without injecting moisture leads to reduction of the NO formation in the flame front. With the injection of a drop of H_2O, the maximum local temperature at any α decreases, and the content of nitrogen monoxide also permanently decreases. The dependencies are close to linear.

As the experimental studies demonstrated, injection of droplet moisture into the flame front ensured the reduction of maximum local temperature in the droplet trace by 200±10 K. This led to a decrease in the nitrogen monoxide content in the combustion products by 40 ... 50%. The most effective is the reduction of NO formation due to the injection of droplet moisture into the flame front at lower α (close to 1.0), i.e. at higher flame temperatures.

The residence time of the reacting substances in the flame front is insignificant (in experimental studies $\tau \approx 2.0{\times}10^{-3}$ s) and, therefore, reactions taking place in the flame front cannot provide equilibrium concentrations of reaction products. In this case, the NO concentration should be primarily a function of the maximum local temperature in the measurement zone. In order to check this assumption, the dependence of the maximum NO concentration in the flame front on the maximum temperature was built with using data obtained at various values of

α and with the injected water droplet (Figure 5, curve 1), as well as of the rate of change of the NO content with temperature (Figure 5, curve 2).
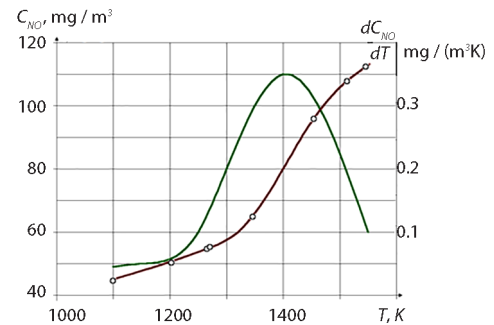

Figure 5. Concentration and intensity of the NO formation in the laminar flame of natural gas: 1 – $\bigcirc$ – NO concentration; 2 – — – intensity of NO formation

For the experimental conditions the most rapid increase in the NO formation occurs in the temperature range of 1350–1500 K, and the rate of change in the NO concentration reaches 0.29 mg/m^3 per degree of temperature. The obtained result indicates that in the laminar flame front the amount of NO formed is determined mainly by the maximum local temperatures and less depends on the method of lowering the temperature: by increasing α or due to the injection of moisture.

2.2. The effect of the addition of hydrogen peroxide on the process of oxidation of NO to NO_2

In order to determine the effect of the peroxide radical HO_2^- on the kinetics of the process of the oxidation of NO to NO_2 during combustion, a series of experiments were carried out, the results of which are shown in Figure 6.
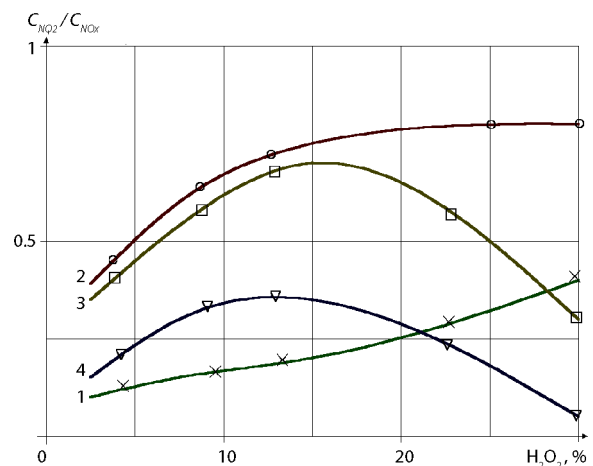

Figure 6. The effect of the oxidizing agent (hydrogen peroxide) concentration on the NO_2 formation in the front of the laminar flame of natural gas: x – 1 – pre-flame zone, coordinate $\delta = -1.0$ mm; $\bigcirc$ – 2 – front edge of the flame front, coordinate $\delta = 0.0$ mm; $\square$ – 3 – mid section of the flame front, coordinate $\delta = 1.0$ mm; ∇ – 4 – back edge of the flame front, coordinate $\delta = 2.0$ mm

Hydrogen peroxide H_2O_2 solution of different concentrations (up to 30%) was injected in the form of droplet moisture into the laminar flame, with using the experimental method described above.

Separate measurements of NO and NO_2 have demonstrated that dependence of the fraction of NO_2 on the concentration of the H_2O_2 solution injected into the flame front is ambiguous and heavily depends on the place of sample taking in the cross section of the flame front. In the low-temperature pre-flame zone, at the coordinate δ = −1.0 mm (Figure 6, curve 1), i.e. 1.0 mm along the normal afore of the edge of the flame front, the portion of NO_2 in the total amount of nitrogen oxides begins to rise monotonously with increasing concentration of H_2O_2 solution injected into the flame front, according to a dependence close to linear. In the initial edge front section (at the coordinate δ = 0.0 mm (just at the very beginning of the temperature rise), curve 2), the NO_2 portion reaches 80% of the maximum at 15% concentration of the H_2O_2 solution and comes to the limit values, and then remains unchanged with any further increase in the concentration of the hydrogen peroxide solution. In the mid section of the flame front (curve 3), an increase in the concentration of H_2O_2 up to 15% also leads to a significant, up to 80%, increase in the portion of NO_2, but the further increase in the percentage of hydrogen peroxide leads to a decrease in this portion to 0.3, which approximately corresponds to the influence of 3% solution of H_2O_2. It means that in the zone of maximum temperatures, the active substance is no longer the peroxide radical HO_2^-, but OH^- hydroxyl takes this place instead, and due to the combustion of natural gas (CH_4) an acute lack of hydrogen is no longer observed in this zone.

Curve 4, corresponding to the section at the coordinate δ = 2.0 mm, qualitatively repeats the previous one, however, with a less pronounced extremum at the point with the coordinate corresponding to 15% of the H_2O_2 concentration. Thus, the temperature above 800 K significantly affects the reaction rates and changes the kinetics of the processes involving hydrogen peroxide, and in different sections of the front these changes are ambiguous, which is apparently due to a decrease in the H_2O_2 lifetime in these temperature zones to 20 ms and less. It is likewise significant that the degree of influence of hydrogen peroxide on the process of the further oxidation of NO to NO_2 for different temperatures has a certain optimum in the concentration of the injected substance, i.e. apparently the point where thermodynamics once again begins to prevail over the kinetics. As can be seen from Figure 6, for these conditions this optimum corresponds to a 15% solution of H_2O_2.

Conclusions

1. The water vapor is demonstrated to be the third very influential participant, along with fuel and oxidizer, in the combustion process of a hydrocarbon fuel.

2. The concept of the "excess moisture ratio" in the combustion process has been introduced, as the ratio of all moisture injected into the process with fuel and oxidant, to that which dissociates and participates in the process chemically.

3. Injection of moisture into the combustion zone of natural gas is demonstrated to have an effect due to the insertion of additional quantities of HO_2^- and OH^- radicals into the process, contributing to the intensification of the further oxidation of NO to NO_2. It is experimentally confirmed that the HO_2^- radical plays a crucial role in the oxidation of NO to NO_2 in the flames of natural gas; at temperatures below 673 K, NO is further oxidized to NO_2 with the peroxide radical HO_2^- as the initiator of the process in the low-temperature post-flame zone.

4. With the injection of moisture into the flame, the temperature in the flame core decreases and the content of the HO_2^- radical increases, and so does the efficiency of oxidation of NO to NO_2 in the post-flame zone.

The maximum decrease in the concentration of NO (by about 50%) in the combustion products was observed when moisture was injected into the process at burning under α close to 1 (α = 1.02).

5. The most favorable condition for accelerating the process of the further oxidation of NO to NO_2 is injection of the 15% solution of hydrogen peroxide.

6. Injection of moisture into the combustion process has a significant effect on the formation of "fast" ("kinetic") nitrogen oxides, and not only on the formation of "thermal" nitrogen oxides (possible influence on which is limited by the Frank-Kamenetsky temperature step), and as a result, can cause the reduction of nitrogen oxide emissions by up to 35%, additional to the thermodynamic effects (usually max 50%).

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