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## **Formation Mechanisms and Methods for Calculating Pollutant Emissions from Natural Gas Combustion Depending on the Burner Emission Class**

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**Abstract.** The combustion of hydrocarbon fuels in the chambers of heat generating plants is one of the main sources of pollutant emissions. Environmental standards and rules that limit emissions are becoming more stringent and their implementation requires the introduction of advanced technologies and equipment. The main device in combustion systems are blow burners, the design of which largely determines the level of emission. The article considers factors that intensify the formation of normalized pollutants, provides global chemical reactions, various types of mechanisms, and kinetic schemes. Based on the analysis of modern methods for reducing harmful emissions, the most effective design solutions for mixing devices, nozzles and systems for distributing the flow of fuel and air supplied to combustion are determined. A comparative analysis of the methods and conditions for determining the emission class of the burner device is carried out depending on the selected units of measure, the coefficient of excess air (oxygen concentration in flue gases), air humidity and the initial composition of natural gas using examples of EU and EAC standards. The methodology for calculating the emissions of nitrogen oxides depending on the measurement conditions is given. The conversion factors for the values of pollutant emissions from the accepted units in the EU (mg/(kW·h)) into the units indicated according to the EAC environmental rules (mg/m<sup>3</sup>) taking into account the respectively normalized coefficient of excess air are obtained. As a result of the calculations, the types of burners were determined by emission classes corresponding to the applicable environmental standards and rules in the Republic of Belarus, depending on the heat output of the boiler plants.

**Keywords:** environmental standards, excess air coefficient, mixing device, flame head, conversion factors, concentration of nitrogen oxides

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## **Механизмы образования и методика расчета выбросов загрязняющих веществ при сжигании природного газа в зависимости от эмиссионного класса горелок**

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**Реферат.** Сжигание углеводородного топлива в камерах сгорания теплогенерирующих установок – один из основных источников выбросов загрязняющих веществ. Экологические

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нормы и правила, ограничивающие выбросы, становятся все более жесткими, и их соблюдение требует внедрения передовых технологий и оборудования. Основным устройством в системах сжигания являются дутьевые горелки, от конструкции которых во многом зависит уровень эмиссии. В статье рассмотрены факторы, интенсифицирующие образование нормированных загрязняющих веществ, приведены глобальные химические реакции, различные типы механизмов и кинетические схемы. На основе анализа современных методов снижения вредных выбросов определены наиболее эффективные конструкторские решения смесительных устройств, насадок и систем распределения потоков топлива и воздуха, подаваемого на горение. Проведен сравнительный анализ методов и условий определения эмиссионного класса горелочного устройства в зависимости от выбранных единиц измерения, коэффициента избытка воздуха (концентрации кислорода в дымовых газах), влажности воздуха и исходного состава природного газа на примерах стандартов ЕС и ЕАС. Приведена методика расчета выбросов оксидов азота в зависимости от условий измерения. Получены коэффициенты пересчета значений выбросов загрязняющих веществ из принятых единиц в ЕС (мг/(кВт·ч)) в единицы, указанные по экологическим правилам ЕАС (мг/м<sup>3</sup>) с учетом соответственно нормируемого коэффициента избытка воздуха. В результате расчетов определены типы горелок по эмиссионным классам, соответствующим действующим экологическим нормам и правилам в Республике Беларусь в зависимости от тепловой мощности котельных установок.

**Ключевые слова:** экологические нормы, коэффициент избытка воздуха, смесительное устройство, пламенная голова, коэффициенты пересчета, концентрация оксидов азота

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## Introduction

With the coming into force in the Republic of Belarus since October 1, 2017 of environmental norms and rules of EcoNiP 17.01.06-001–2017 “Environmental protection and nature management. Environmental safety requirements” [1], the issue of limiting harmful emissions not only in existing boiler houses, the emission standards of which are set slightly higher than for newly built ones, became acute, and a reduction in existing indicators can be resolved by installing condensing heat exchangers at the exit from the boilers and improving heat and mass transfer processes in boiler furnaces [2], but also for newly designed boiler plants, emission standards for which are significantly tightened.

In this regard, for manufacturers of boilers, the task is to optimally design newly manufactured plants, and for design organizations – a quality choice of equipment offered on the market. The complexity of the solution of this problem is primarily due to the fact that the manufacturers of burner devices uniquely determine the emission class of their products based on the measured values when burning the corresponding types of fuel in once-through furnaces significantly exceeding the size of the free flame, having an extremely low aerodynamic drag and almost complete absence of reverse flows [3]. Boilers with similar combustion chambers, usually referred to as single-pass or span, due to the need to retrofit a particular system of utilizing the heat of the flue gases to increase the overall efficiency of the installation [4], are rarely used and mainly as energy

ones. In the market of heating and industrial boilers, currently mainly boilers with two-way reversible and three-way continuous combustion chambers are offered. An additional difficulty in unambiguously determining the level of emissions in accordance with [1] is the discrepancy in the accepted conditions and units for measuring the concentration of pollutants in the EU [3] – the main producer of burner devices, as well as incomplete compliance of the chemical composition and, as a result, the composition of flue gases with standard types of fuel in the EU and EAC. The proposed simplified conversion methods, for example, described in [5, 6], can lead to design inaccuracies and, as a result, to errors in choosing the optimal designed equipment. For more accurate calculations, it is necessary to apply a combined technique, taking into account the described factors.

### **Main part**

Before considering the features of the effect of the emission class of burners on the formation of harmful emissions, it is necessary to determine the actual composition of the flue gases and which pollutants should be determined as the object of study. According to [1], when burning gaseous fuels, the following issues are standardized: carbon oxide (CO), nitrogen oxides – in terms of nitrogen dioxide (NO<sub>x</sub>), sulfur dioxide (SO<sub>2</sub>); and when burning liquid fuel, the same substances plus solid particles.

The factors that intensify the formation of each of the normalized pollutants are considered below.

Carbon monoxide is formed, primarily and almost exclusively, by the combustion of fossil fuels due to the incomplete oxidation of hydrocarbon molecules [7]. To reduce its amount in flue gases, the flow rate of air entering the combustion is increased. It is with a decrease in the formation of this gas that such a concept as the coefficient of excess air is associated. However, the desire of many installers of gas burner devices to completely get rid of CO by increasing the volume of air supplied to the combustion leads to an overestimation of the excess air coefficient and, as a result, to a decrease in the technical efficiency of the heat generating unit and to some extent an increase in the emission of another normalized pollutant – nitrogen oxides. We also note that, according to experimental studies [8], the process of converting hydrocarbon fuels to the final products of combustion of H<sub>2</sub>O and CO<sub>2</sub> is divided into two stages: the first is the oxidation of hydrocarbons to CO – the rate of processes in a high-temperature medium (above 600 °C) is very high; the second is slow: oxidation of CO to CO<sub>2</sub>. Proceeding from this, the qualitative oxidation of CO depends not only on temperature, but to a large extent on the time spent in the high-temperature zone. This conclusion is extremely important when comparing processes occurring in reversible and continuous combustion chambers. Under conditions of reverse flue gas flows, not only does the average molecular path inside the furnace increase, but their speed also slows down due to cross-border turbulent interpenetrations of multidirectional peripheral flows, viz. a flow of a bur-

ning air-fuel mixture and a return flow of combustion products [9]. As a result, this leads to a significantly longer (almost 2 times) period of time for the oxidation of CO directly in the furnace compared to through-passage combustion chambers. During commissioning, which consists in fixing certain fuel-air ratios in an adjustable power range, presented both by the boiler manufacturer and the burner manufacturer for each type of device, such a mechanism, as a result, can significantly reduce the excess air required for CO oxidation to normalized values. In this case, the coefficient of excess air will weakly depend on changes in the power of the heat generating unit. In the passage furnaces, where the time allocated for CO oxidation is almost proportional to the flow rate, the excess air coefficient will increase with increasing power for a given furnace size. However, on the other hand, modern boilers, unlike earlier ones, have so-called “long furnaces” and CO oxidation to normalized values occurs already when the amount of O<sub>2</sub> in the exhaust gases is ~ (3–3.5) %, which corresponds to a quite acceptable excess air coefficient up to 1.2. It should be also noted that in the table. E10 [1] for boiler plants with a rated capacity of more than 0.1 MW commissioned on January 1, 2019, the standards for carbon monoxide emissions are not standardized at all – up to a plant capacity till 25 MW.

Nitrogen oxides and, above all, nitrogen monoxide during the combustion of fuels containing a small amount of bound nitrogen, are formed mainly in the high temperature zone of ~1850 °C according to the so-called “Zeldovich mechanism” [10]:



which subsequently [11] was added by the reaction



Together these reactions are usually called the “extended Zeldovich mechanism”. However, it was noted that the experimentally measured concentrations of NO<sub>x</sub> in the exhaust gases exceed those calculated by the Zeldovich mechanism. An explanation of the additional mechanism of the formation of nitrogen oxides is associated with the presence of the CH radical in the initial combustion zone, which reacts with molecular nitrogen [12]:



These reactions are called by the name of their discoverer “Fenimore mechanism” or, in association with their occurrence almost exclusively in the initial combustion zone, – “fast mechanism”. Currently, it is believed that NO is formed from NCN in a number of subsequent reactions involving various radicals [13]. Given the individual reactions defined in [11–13] for the general picture of the process, we present a generalized kinetic diagram of the formation of NO by the fast mechanism (Fig. 1).

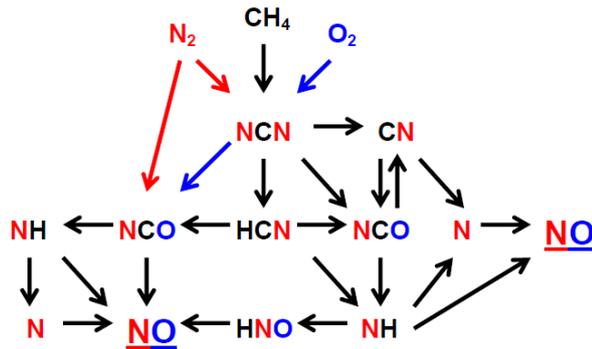


Fig. 1. The scheme of reactions for NO formation via the prompt-NO mechanism

Since reactions according to the “fast mechanism” occur in the initial combustion zone, to determine their contribution to the total  $\text{NO}_x$  concentration in the flue gases, the simplest and most realistic approach is to partially exclude the influence of the thermal mechanism with subsequent measurements of the actual  $\text{NO}_x$  values. The organization of poor mixtures burning almost completely eliminates the thermal mechanism, especially when burning of natural gas. Therefore, using this method, it is possible to determine the concentration of nitrogen monoxide formed by the “fast mechanism”, and it is widely used in gas turbine plants [14]. For its implementation, a pre-prepared poor fuel-air mixture (with a significant excess of the excess air coefficient) is fed into the combustion chamber. Moreover, due to excess ballast gas, the temperature of the combustion products does not reach the values required for the reactions of the Zeldovich mechanism. However, to reduce the total concentration of nitrogen oxides, it is precisely the part formed by the “fast mechanism” that is least affected by the applied external devices that organize mixing cooling flows and/or flame separation, due to their occurrence exclusively in the initial combustion zone. In addition to the Zeldovich and Fenimore mechanisms described above, a low-temperature mechanism [15] related to the decomposition of nitrogen-containing fuel components can make a significant contribution to the total concentration of  $\text{NO}_x$  in flue gases. But the types of fuel that are commonly used with blast burners – natural gas and light liquid fuels – usually have a very small amount of nitrogen in their composition, and this mechanism seems to be important in the pyrolysis and direct combustion of solid fuels and secondary fuels of chemical plants. In any case, the design of the burner devices cannot significantly reduce the formation of nitrogen oxides obtained as a result of the action of fast and low-temperature mechanisms. For these mechanisms, the most effective way to reduce  $\text{NO}_x$  in exhaust gases seems to be their purification. For this, the most widespread means are: selective catalytic [16] and selective non-catalytic [17] recovery, the implementation of which requires additional sophisticated equipment and significant capital costs. Thus, in order to ensure normalized  $\text{NO}_x$  values in the flue gases of heating and industrial boilers when working with blow burners, it is first necessary to consider the factors affecting the rate of reactions according to the Zeldovich mechanism, since this is the only mechanism that

can be influenced by optimizing internal flows in the combustion chamber, and also due to the fact that the reactions proceeding along this path make the most significant contribution to the formation of nitrogen oxides in the high-temperature zone. In order to do this, we divide this task into two, viz. determining effective factors for the design of burner devices, on the one hand, and the design of combustion chambers, on the other, and then combine them to determine a qualitative assessment of the mutually affecting processes and characteristics. The first part of this task will be considered in the present article.

Blow gas burner devices according to the generation of pollutant emissions are divided into three emission classes [3, 18]: 1<sup>st</sup> class –  $\text{CO} \leq 120 \text{ mg}/(\text{kW}\cdot\text{h})$ ,  $\text{NO}_x \leq 170 \text{ mg}/(\text{kW}\cdot\text{h})$ ; 2<sup>nd</sup> class –  $\text{CO} \leq 80 \text{ mg}/(\text{kW}\cdot\text{h})$ ,  $\text{NO}_x \leq 120 \text{ mg}/(\text{kW}\cdot\text{h})$ ; 3<sup>rd</sup> class –  $\text{CO} \leq 60 \text{ mg}/(\text{kW}\cdot\text{h})$ ,  $\text{NO}_x \leq 80 \text{ mg}/(\text{kW}\cdot\text{h})$ .

Currently approved methods of reducing emissions from the combustion of hydrocarbon fuel are:

- 1) minimizing the coefficient of excess air to ensure complete combustion of fuel;
- 2) the introduction of cooling flows into the combustion zone (recirculation of part of the flue gases; injection of steam, water, etc.);
- 3) two-stage combustion of fuel (creation of a primary and secondary flame);
- 4) distribution of fuel to the periphery of the flame (creating a group of peripheral flames with the smallest volume of flame nuclei);
- 5) reducing the temperature of the heating of the air entering the combustion.

The last method is a regime-technological one and cannot be implemented solely due to the design of the burner. Briefly considered are the remaining methods.

1. A low coefficient of excess air can be ensured by high-quality mixing of the fuel with the flow of air forced into the combustion. For this, various kinds of mixing devices are used, as a rule, twisting and dividing the gas-air mixture flow into a number of smaller flows. This approach allows one to obtain high-quality combustion with a slight excess of air ( $\lambda \sim (1.15-1.17)$ ) and practically

solve the problem of reducing CO emissions to minimum values. Such a burner design may well provide emissions in the 2<sup>nd</sup> emission class (Fig. 2). However, to reduce the generation of NO molecules to guaranteed values of the 3<sup>rd</sup> emission class, this method is not enough.

2. The introduction of additional flows of external media (steam, water) is associated with an additional complication of the entire system and, as a consequence, a significant increase in the cost not only of the burner device and the external cooling medium supply system, its significant complexity and the growth not only of capital (equipment



*Fig. 2.* Mixer head of the 2<sup>nd</sup> emission class burner. Gas burner MG3 designed by Enertech GmbH Division Giersch (Germany)

cost), but also operating costs. As a result, the main direction of application of this method is the creation of recirculating flows of flue gases into the combustion zone. Such flows can be created by additional external caps on the flame tube of the burner (Fig. 3) or by a device for exiting the air-fuel flow from the flame tube (for example, narrowing the flow with an external ring using the Coande effect [19]) (Fig. 4).

3. The creation of a two-stage flame not only significantly complicates the combustion system, but also reduces the range of regulation of the power of the burner in conditions of stable complete combustion of fuel. And yet the generation of interdependent flames requires additional sensors and an interdependent regulation system. Given that modern requirements for heat generators include a wide range of power modulation, this method can be primarily effective in systems with stable heat consumption, which significantly limits its application. However, it should be noted that the stepped flames is an effective method for simultaneous combustion of several types of fuel in multi-fuel burners [20–22].



Fig. 3. Various types of caps on the flame tube of the burner to create flue gas recirculation.  
Photo from the test laboratory of Enertech GmbH Division Giersch (Germany)



Fig. 4. Flame head of a burner of the 3<sup>rd</sup> emission class with a nozzle providing recirculation of flue gases with the formation of the Coandă effect. Gas burner MG3-LN designed by Enertech GmbH Division Giersch (Germany)

4. The method of distributing the hottest combustion zones (nuclei) to the periphery of the flame is one of the most effective ones and widely used in modern burner devices. For its implementation, it is enough to divide the fuel flow into several independent jets directed to the periphery of the flame (Fig. 5).

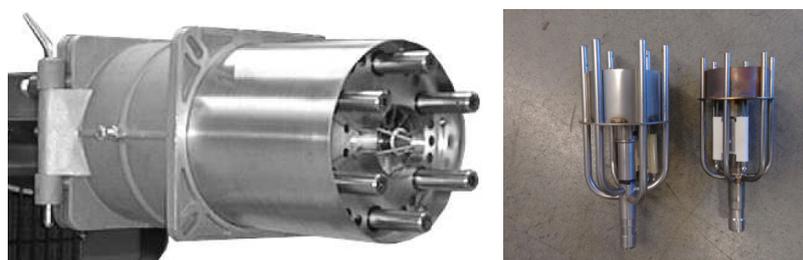


Fig. 5. Flame head and distribution nozzles of the 3<sup>rd</sup> emission class burner with gas flow fission system. Gas burner MG10-LN designed by Enertech GmbH Division Giersch (Germany)

Thus, the main methods for providing the 3<sup>rd</sup> emission class are reduced to the use of burner designs according to the 2<sup>nd</sup> and 3<sup>rd</sup> of the described methods, or a combination thereof.

It should be noted that burner manufacturers according to the EN DIN standard [3] determine the quantitative values of the concentration of pollutants at the outlet of the combustion chamber (in mg/(kW·h)) for dry gases with air humidity entering the combustion  $d = 10$  g/kg [23]. These units (mg/(kW·h)) are selected because, when used, the amount of pollutants refers to the unit of generated heat energy and, in this case, it does not matter during stoichiometric or non-stoichiometric combustion measurements are made, i. e. the amount of O<sub>2</sub> in the flue gas is not required. At first glance, such an approach seems correct. Indeed, with an increase in excess air, the volume of emissions increases, but the average temperature of the exhaust gases decreases. The heat capacity of CO<sub>2</sub> [24] and H<sub>2</sub>O [25] – the main components of the flue gases – varies slightly in the temperature range of stable combustion, and the amount of heat, defined as the product of the average temperature and volume, remains close to unchanged for different values of the coefficient of excess air. In the Republic of Belarus and the CIS countries, emission indicators are determined (in mg/m<sup>3</sup>). Such units are directly dependent on the volume of emissions. As a result, the problem arises of unambiguous conversion of these units. For this, it is necessary to set an additional value – either by the coefficient of excess air  $\lambda$ , or by the volume concentration of oxygen in the flue gases  $K_{V_{O_2}}$ . Because high-quality combustion of the air-fuel mixture by modern blast burners with the lowest heat loss with flue gases is possible at  $\lambda \sim (1.15-1.20)$ , which corresponds to an oxygen concentration of  $K_{V_{O_2}} \sim (2.8-3.5)$  %, then the burner manufacturers have established a convenient conversion rule for the average whole fixed value  $K_{V_{O_2}} = 3$  % [5]. But for complete uniqueness, it is also required to have a coefficient of direct conversion of mg/(kW·h) to mg/m<sup>3</sup>. For this, the inverse coefficient  $f$  is usually indicated, the value of which varies according to different sources, for example,  $f = 1.001$  [5] or  $f = 1.164$  [26] for natural gas of class E (H) and  $f = 1.018$  for natural gas of class L (LL) [5] at  $K_{V_{O_2}} = 3$  % or  $f = 0.857$  at  $K_{V_{O_2}} = 0$  % [27]. The conditions

for finding the conversion factor are not specified. Moreover, the empirically convenient, but illegitimate for direct calculations, setting  $K_{V_{O_2}} = 3\%$  in the information and reference catalogs of burner manufacturers [5] leads to obtaining not quite accurate calculated values of conversion factors. This is due, first of all, to the fact that in European countries mixed combustible gases of various origins are very often used. Therefore, according to the EU standard [3], combustible gases are not standardized by the calorific value or constituent components, but by the Wobbe number [28]:

$$W_i = \frac{H_{i,n}}{\sqrt{d}}, \quad (4)$$

where  $H_{i,n}$  – net calorific value, MJ/m<sup>3</sup>;  $d = \frac{\rho_{gas}}{\rho_{air}}$  – relative density of air;

$\rho_{gas}$ ,  $\rho_{air}$  – density of gas and of air under normal conditions, kg/m<sup>3</sup>.

The Wobbe number is an indicator of gas quality. Using this criterion, one can verify the interchangeability of fuel gases. So, the same Wobbe number means the constancy of the heat flux at the same pressure of the supplied gas, regardless of its composition. For example, for a practitioner it is absolutely necessary in case of a change in gas supply to establish with the of the Wobbe number whether the rated load is still guaranteed [6].

The LL group (previously called L – Low) includes combustible gases with the Wobbe number:  $W_i = 10.5\text{--}13.0$  kW·h/m<sup>3</sup>. The Wobbe number equal to  $W_i^0 = 12.4$  kW·h/m<sup>3</sup> is taken as the nominal value for this group of combustible gases. The second group E (previously called H – High) includes combustible gases with  $W_i = 12.8\text{--}15.7$  kW·h/m<sup>3</sup>. The nominal value for this group is  $W_i^0 = 15.0$  kW·h/m<sup>3</sup> [6]. Given that the manufacturers of burners, mainly used in the CIS, are from different European countries and, above all, from Germany, they apply all calculations, operating parameters, indicate the working zones of the burners, etc., based on the types of natural gas used in these countries. In this case, the net calorific value  $H_{i,n}$  of gas E (H) is given in the range of 35.32–37.44 MJ/m<sup>3</sup>, and that of LL (L) gas – is 30.12–31.77 MJ/m<sup>3</sup>. Thus, natural gas that is used in the CIS countries both by Wobbe number and by calorific value can be identified by European standard as LL gas. According to [29], in the CIS countries, the net calorific value of natural gas is not less than 31.80 MJ/m<sup>3</sup>. As a result, being guided by the values of the calorific value, the designers and adjusters of the blast burners use the data for the LL (L) group for power characteristics, bearing in mind that the real natural gas entering our boiler houses is at least no less caloric. But how appropriate is it to apply these data when calculating emissions? After all, emissions depend not only on the calorific value of the gas, but also on its composition. The Tab. 1 shows the compositions of several real natural gases and the corresponding calculated coefficients  $f$ .

Table 1

**Compositions of typical natural gases and their characteristics  
needed to convert the concentration of emissions from mg/(kW·h) to mg/m<sup>3</sup>**

Name	Chemical formula	Unit	Gas LL (L) acc. to [6]	Gas E (H) acc. to [6]	Russian natural gas acc. to [30]	Russian natural gas acc. to [31]
			1	2	3	4
Methane	CH <sub>4</sub>	volume %	81.8	92.3	97.04	98.0
Ethane	C <sub>2</sub> H <sub>6</sub>	volume %	2.8	2.0	0.99	0.5
Propane	C <sub>3</sub> H <sub>8</sub>	volume %	0.4	1.0	0.381	0.3
I-butane	CH(CH <sub>3</sub> ) <sub>3</sub>	volume %	0.1	0.3	0.0516	0.05
H-butane	CH <sub>3</sub> -CH <sub>2</sub> - -CH <sub>2</sub> -CH <sub>3</sub>	volume %	0.1	0.3	0.06	0.05
I-pentane	(CH <sub>3</sub> ) <sub>2</sub> -CH- -CH <sub>2</sub> -CH <sub>3</sub>	volume %	-	-	0.0136	0.1
H-pentane	CH <sub>3</sub> -(CH <sub>2</sub> ) <sub>3</sub> - -CH <sub>3</sub>	volume %	-	-	0.0106	0.1
Neo-pentane	(CH <sub>3</sub> ) <sub>4</sub> C	volume %	-	-	0.00065	-
Hexanes	C <sub>6</sub> H <sub>14</sub>	volume %	-	-	0.0074	-
Nitrogen	N <sub>2</sub>	volume %	14.0	3.1	1.29	0.8
Carbon dioxide	CO <sub>2</sub>	volume %	0.8	1.0	0.127	0.1
Oxygen	O <sub>2</sub>	volume %	-	-	0.0148	-
Gas density	$\rho_{gas}$	kg/m <sup>3</sup>	0.829	0.784	0.6891	0.674
Lower calorific value	$H_{i,n}$	MJ/m <sup>3</sup> (kW·h/m <sup>3</sup> )	31.80 (8.83)	36.00 (10.00)	33.52 (9.31)	32.60 (9.06)
The value of the Wobbe number, cal- culated acc. to (4)	$W_i$	MJ/m <sup>3</sup> (kW·h/m <sup>3</sup> )	39.75 (11.04)	46.24 (12.84)	40.38 (11.22)	40.07 (11.13)
Stoichiometric air volume calculated acc. to (5)	$V_{air}^T$	m <sup>3</sup>	8.496	9.773	9.600	9.620
Air volume at O <sub>2</sub> = 3 %, calcula- ted acc. to (6)	$V_{air}^R_{at\_O_2=3\%}$	m <sup>3</sup>	9.915	11.41	11.20	11.23
The moisture volu- me at O <sub>2</sub> = 3 %, cal- culated acc. to (7)	$V_{H_2O}^{O_2=3\%}$	m <sup>3</sup>	1.905	2.123	2.172	2.184
The moisture volume at O <sub>2</sub> = 0, calculated acc. to (7)	$V_{H_2O}^{O_2=0\%}$	m <sup>3</sup>	1.882	2.096	2.147	2.158
The volume of dry gases at O <sub>2</sub> = 3 %, calculated acc. to (8)	$V_{dry\_gas\_at\_O_2=3\%}^R$	m <sup>3</sup>	9.01	10.29	10.03	10.05
The volume of dry gases at O <sub>2</sub> = 0, calculated acc. to (8)	$V_{dry\_gas\_at\_O_2=0\%}^R$	m <sup>3</sup>	7.61	8.68	8.45	8.46
The conversion fac- tor mg/m <sup>3</sup> into mg/(kW·h) at O <sub>2</sub> = 3 %, calculated acc. to (9)	$f_{O_2=3\%}$	-	1.074	1.029	1.077	1.110
The conversion fac- tor mg/m <sup>3</sup> into mg/(kW·h) at O <sub>2</sub> = 0, calculated acc. to (9)	$f_{O_2=0\%}$	-	0,861	0.868	0.907	0.935

For natural gases, the theoretically necessary volume of combustion air can be calculated using the reduced formula

$$V_{air}^T = \frac{1}{V_{O_2}^0} \left[ \sum \left( m + \frac{n}{4} \right) C_m H_n - O_2 \right], \quad (5)$$

where  $V_{O_2}^0 = 20.946$  – volume concentration of oxygen in atmospheric air [32];  $C_m H_n$  – hydrocarbons that are part of the gas;  $m, n$  – hydrocarbon indices for carbon and hydrogen, respectively;  $O_2$  – volume concentration of oxygen in a combustible gas.

For technological and artificial combustible gases where CO, H<sub>2</sub> and H<sub>2</sub>S are present:

$$V_{air}^T = \frac{1}{V_{O_2}^0} \left[ 0.5(CO + H_2) + 1.5H_2S + \sum \left( m + \frac{n}{4} \right) C_m H_n - O_2 \right]. \quad (5')$$

The manufacturers of blow burners, as was already mentioned, determine the concentration of pollutant emissions with residual oxygen  $K_{V_{O_2}} = 3\%$ , which corresponds to  $\lambda = 1.167$ . But the essence of the problem is not to determine the conversion factor for a given specific value of oxygen in the flue gas, but to determine the limiting emission indicators for a particular emission class of burners. Although the readings in mg/(kW·h) really practically do not depend on the amount of oxygen in the flue gases, yet, since with an increase in excess air, the volume of exhaust gases will increase, the conversion factor in mg/m<sup>3</sup> will increase, too. If we need to determine the conversion factor for the residual oxygen content in flue gases  $K_{V_{O_2}} = X\%$ , then the calculation should be attributed to the volume of combustion air increased by the excess air coefficient

$$V_{air\_at\_O_2=X\%}^R = V_{air}^T \frac{V_{O_2}}{V_{O_2} - K_{V_{O_2}}} = \lambda V_{air}^T. \quad (6)$$

After finding this value, it would seem that it is possible to calculate the conversion factor of thermal units into volume units. However, if the standard gas analyzers used, so that the moisture of the flue gases (and NO<sub>2</sub> is dissolved in it, which gives it a certain degree of aggressiveness) from the sample taken does not fall on the sensitive element, but is condensed and drained. As a result, the readings of the gas analyzer relate to dry flue gases. Therefore, it is necessary to calculate the volume of dry combustion products. For this, it is more convenient to first find the amount of moisture in the flue gases:

$$V_{H_2O}^{O_2=X\%} = 0.01 \left( \sum \frac{n}{2} C_m H_n + 0.1 \frac{\rho_{air}^0}{\rho_{H_2O}} RHV_{air\_at\_O_2=X\%}^R \right), \quad (7)$$

where  $\rho_{air}^0 = 1.293$  kg/m<sup>3</sup> – density of dry air under normal conditions;  $\rho_{H_2O}^0 = 0.840$  kg/m<sup>3</sup> – density of water vapor under normal conditions;

$RH$  – relative humidity, g/kg; 0.01 – coefficient recalculation of the content of ingredients % → share;  $0.01 \cdot 0.1 = 0.001$  – conversion factor kg → g.

Since we determined the specific theoretical volume of air for combustion, we will add 1 m<sup>3</sup> (i. e. the volume of combustible gas) to find the specific volume of flue gases. Then the volume of dry combustion products

$$V_{dry\_gas\_at\_O_2=X\%}^R = (V_{air\_at\_O_2=X\%}^R + 1) - V_{H_2O}^{O_2=X\%}. \quad (8)$$

For calculation, we accept the conditions of relative humidity in accordance with those adopted in [5]:  $RH = 10$  g/kg. As a result, to convert mg/(kW·h) to mg/m<sup>3</sup>, we find the inverse coefficient

$$f = 3.6 / (H_{i,n} / V_{dry\_gas\_at\_O_2=X\%}^R), \quad (9)$$

where 3.6 – conversion factor kW·h → MJ.

Using this method of conversion, it is possible to obtain the values of pollutant emissions in mg/m<sup>3</sup>, if the limiting values are indicated in mg/(kW·h), but it is specifically stipulated at what value of the residual oxygen or excess air coefficient. Indeed, when confirming the certificate of emission class for a particular type of burner, qualification tests are carried out with an excess of air for complete combustion of fuel. For a high-quality burner, this excess will amount to ~3 % oxygen in flue gases. And since mg/(kW·h) refers to the heat released, then the concentration values in these units will practically not depend on the oxygen content. Therefore, no conversion to stoichiometric parameters is required, and burner manufacturers justifiably approve those oxygen values that were measured during qualification tests.

As a result, it is possible to determine the limit values of harmful emissions in mg/m<sup>3</sup> for a particular type of burner that have received the European classification in mg/(kW·h) for a specific composition of combustible gas, substituting  $K_{V_{O_2}} = 3$  % in formulas (6)–(9). As an example, we cite Tab. 2 correspondence of emission classes of burners for natural gas compositions specified in Tab. 1.

Table 2

Limit values of pollutants for gas burners at  $K_{V_{O_2}} = 3$  %

Emission class of burners acc. to [3]	CO limit value					NO <sub>x</sub> limit value				
	mg/(kW·h)	mg/m <sup>3</sup>				mg/(kW·h)	mg/m <sup>3</sup>			
		1	2	3	4		1	2	3	4
1 <sup>st</sup>	120	112	117	111	108	170	158	165	158	153
2 <sup>nd</sup>	80	74	78	74	72	120	112	117	111	108
3 <sup>rd</sup> (Low-NO <sub>x</sub> )	60	56	58	56	54	80	74	78	74	72

It should be noted once again that according to [3] the indicated limit values relate to dry exhaust gases reduced to normal physical conditions, with a relative humidity of 1 % (10 g/kg) and an oxygen content of 3 % (excess coefficient

of air 1.167). In current standards [1] relative air humidity is not mentioned at all, and the coefficient of excess air when calculating emission standards for boiler plants with a capacity of more than 0.1 MW is indicated as 1.4 (oxygen content in flue gases 6 %) – p. 10.1.3 [1]. Thus, the conversion factor from 3 % oxygen to 6 %, calculated by the formula (6), will be

$$K_{3\% \rightarrow 6\%}^{O_2} = (V_{O_2}^0 - 6) / (V_{O_2}^0 - 3) = 0.833. \quad (10)$$

Then, taking into account, as already mentioned, the relevance of standardization for CO, gas burners of the corresponding emission class will have NO<sub>x</sub> limit values for the composition of natural gases indicated in Tab. 1 recalculated taking into account the coefficient  $K_{3\% \rightarrow 6\%}^{O_2}$ .

Table 3

**Compliance of the NO<sub>x</sub> limit values for gas burners with a capacity of more than 0.1 MW with current standards according to [1] for boiler plants of the corresponding capacity**

Emission class of burners acc. to [3]	NO <sub>x</sub> <sup>O<sub>2</sub>=6%</sup> , mg/m <sup>3</sup>									
	Calculation				The rate of heat output, MW					
	1	2	3	4	0.1–0.3	0.3–2.0	2.0–25	25–50	50–100	100<
1 <sup>st</sup>	132	137	132	127	80	100	120	140	100	100
2 <sup>nd</sup>	93	97	92	90						
3 <sup>rd</sup> (Low-NO <sub>x</sub> )	72	68	72	74						

The 1<sup>st</sup> emission class of burners is not suitable for the limiting emission indicators according to environmental standards of almost all countries, including Belarus. As can be seen from the Tab. 3, burners of the 2<sup>nd</sup> emission class (they are often called: class “standard”) are not suitable for use in boilers with a capacity of 0.1–0.3 MW.

It should be noted that in some cases environmental standards are referred to as stoichiometric parameters, i. e. to theoretically necessary air volume. For example, in p. 10.1.2 [1] – these are the regulation rules for boiler plants with a capacity of up to 0.1 MW. In world practice, for example, in the description of the Building Research Establishment Environmental Assessment Method (BREEAM) developed by the British organization BRE Global [27]; for the conversion of thermal emission units into volumetric units, it is indicated that to determine the emission class devices for the combustion of fossil fuels, the values of NO<sub>x</sub> concentration are determined for dry flue gas in the absence of oxygen in the exhaust flue gas. For these cases, the calculation is carried out according to the same above method, but with an excess of oxygen  $K_{V_{O_2}} = 0\%$ .

Table 1 shows the corresponding calculated values for the selected examples of the composition of natural gases. The corresponding emission limit values for gas burners by emission classes for this case are shown in Tab. 4.

Table 4

**Compliance of the NO<sub>x</sub> limit values for gas burners with forced combustion air supply with a power of less than 0.1 MW to the current standards according to [1] for boiler plants of the corresponding rated power**

Emission class of burners acc. to [3]	NO <sub>x</sub> <sup>O<sub>2</sub>=0%</sup> , mg/m <sup>3</sup>				Norm acc. to [1]
	Calculation				
	1	2	3	4	
1 <sup>st</sup>	<b>197</b>	<b>196</b>	<b>187</b>	<b>182</b>	150
2 <sup>nd</sup>	139	138	132	128	
3 <sup>rd</sup> (Low-NO <sub>x</sub> )	93	92	88	86	

As it can be seen from Tab. 4 and for boiler plants with a capacity of less than 0.1 MW, the first emission class of burners is not suitable for limiting emission indicators according to environmental standards [1].

Now, having clarified the correspondence of the emission classes of the burners, it is necessary to determine what, in fact, is measured by standard gas analyzers, which are mainly used by specialists when setting up and determining the concentration of components in the combustion products. As a rule, the instruments are used for these purposes, the operating principle of which is based on physical analysis methods, including auxiliary physicochemical processes (thermochemical, electrochemical, photoionization, photocolometric, chromatographic, etc.). To determine the concentration of NO<sub>x</sub> in combustion products, the most common electrochemical gas analyzers with the determination of mass concentrations of components in a mixture of gases, which are flue gases. An electrochemical sensor is used as a sensitive element. The analyzed gas medium is passed through a selective filter and a hydrophobic membrane and diffuses onto the measuring electrode. In this case, the released electrons passing through the electrolyte and the reference electrode form a direct current signal in the external circuit, the value of which is proportional to the concentration of the analyzed gas. Selective filters have a limited life. As a result, with each measurement, its time increases and the sensitivity of the sensor decreases. In addition, heating the sensor leads to an increase in the readings of the measured concentration, because temperature, according to Boltzmann's law, is only a statistical expression of the kinetic energy, and, consequently, the mean square velocity of the molecules. As a result, additional electrons can be released – the current in the circuit will increase and, as a result, the instrumental readings of the gas concentration will increase at its really constant value. Thus, to obtain instrumental readings corresponding to the actual concentration of the analyzed gas, a number of conditions must be observed: one is to use selective filters and sensors for a limited number of measurements, to prevent the sensor from heating, carry out measurements only when the combustion mode is steady, to take samples only at a specific point at the outlet from the combustion chamber in the absence of excess pressure. Not exact observance of each of these rules will lead to overestimated readings of the device, but, given the rigidity of modern limit concentration standards, to instrumental (but not always real) excess

of these standards as well. The concentration of the analyzed gas, gas analyzers measure directly in quantitative units [ppm] (parts per million), i. e. in parts of the analyzed gas per one million parts of the test gas mixture. For these units, recalculation into volume units adopted in [1] is carried out directly in the analytical program embedded in the device and is practically unambiguous. Calculations are made for normal physical conditions at 0 °C and 101325 Pa [33]. Thus, the recount is carried out according to the well-known formula [34]

$$X [\text{mg}/\text{m}^3] = (Y [\text{ppm}]) \cdot (\text{molar mass}) / (22.4 [\text{liter}]), \quad (11)$$

where 22.4 – volume value in liters of 1 mole of ideal gas accepted in the calculations (more accurate value: 22.413996 [34]);  $Y$  – measured value of the concentration of the analyzed gas, ppm;  $X$  – calculated value of the concentration of the analyzed gas,  $\text{mg}/\text{m}^3$ .

The result is the following conversion factors for normalized pollutants accurate to the fourth digit (0.01 %):

– the molar mass of CO = 28.01 g/mol, then for CO

$$\text{CO} [\text{mg}/\text{m}^3] = (\text{CO} [\text{ppm}]) \cdot (28.01/22.414) = \text{CO} [\text{ppm}] \cdot 1.2497; \quad (12)$$

– the molar mass of NO<sub>2</sub> = 46.0055 g/mol, then for NO<sub>2</sub>

$$\text{NO}_2 [\text{mg}/\text{m}^3] = (\text{NO}_2 [\text{ppm}]) \cdot (46.0055/22.414) = \text{NO}_2 [\text{ppm}] \cdot 2.0525; \quad (13)$$

– the molar mass NO = 30.0061 g/mol, then for NO

$$\text{NO} [\text{mg}/\text{m}^3] = (\text{NO} [\text{ppm}]) \cdot (30.0061/22.414) = \text{NO} [\text{ppm}] \cdot 1.3387. \quad (14)$$

However, according to the Gay – Lussac law [35], at constant pressure, the volume of the constant mass of gas is proportional to the absolute temperature:  $V_1/T_1 = V_2/T_2$ . Those conversion factors will change at the operating temperature for determining the concentration of the analyzed gas. Thus, it is necessary to compensate for the temperature factor during measurements. In gas analyzers used in practice, an integral resistance with a negative temperature coefficient serves to compensate for the effect of temperature, which ensures the stability of the sensor regardless of temperature [26]. But how much does the law of change in the integrated integral resistance of the gas analyzer coincide with the real law of the dependence of the density of the analyzed gas on temperature? The question remains open, because with increasing temperature and concentration of the analyzed gas, the deviation from the calculation law for an ideal gas will increase (and not linearly). This is especially significant at temperatures close to the dew point temperature with a high concentration of the analyzed gas. For example, the use of the Van der Waals equation [36] as an estimated equation allows an error of up to ~10 %, the Diterichi equation [37] up to ~7 %, the Berthelot equation [38] up to ~5 %, the Redlich – Kwong equation (modifications of Soave) [39] – up to ~(3–4) %. As a result, the value indicated by the device in  $[\text{mg}/\text{m}^3]$  will be a little overestimated.

It should also be taken into account that the so-called “field” gas analyzers, as a rule, do not have an installed NO<sub>2</sub> sensor, and the measured amount of NO is determining the total amount of NO<sub>x</sub> in flue gases. Moreover, the amount of NO<sub>2</sub>, as a rule, is not calculated at all [40]. Recalculation, as a rule, is carried

out only on the total amount of all nitrogen oxides, i. e. on  $\text{NO}_x$ , according to the following programmed formulas:

$$\begin{aligned} \text{NO}_x [\text{ppm}] &= \text{NO} [\text{ppm}] \cdot 1.05 \text{ (without installed NO}_2 \text{ sensor) [40]} \\ \text{or NO}_x [\text{ppm}] &= \text{NO} [\text{ppm}] \cdot (100/97) \text{ (without an installed NO}_2 \text{ sensor) [26];} \\ \text{NO}_x [\text{ppm}] &= \text{NO} [\text{ppm}] + \text{NO}_2 [\text{ppm}] \text{ (with pre-installed NO}_2 \text{ sensor).} \end{aligned}$$

According to the conditions adopted in [1],  $\text{NO}_x$  emissions are normalized in terms of  $\text{NO}_2$ . As a result, to determine the amount of emissions [in  $\text{mg}/\text{m}^3$ ], the obtained value for  $\text{NO}_x$  [in ppm] is programmatically multiplied by the conversion factor for  $\text{NO}_2$ , despite the fact that the main amount of  $\text{NO}_x$  emissions is NO. It was determined that the entire volume of NO in the exhaust gases is oxidized to  $\text{NO}_2$ . And although this reaction is generally reversible, for the reverse action (NO decomposition), sufficiently high temperatures are needed – more than 700 °C [41]. The probability of a reverse process at operating temperatures of flue gases ((120–150) °C) is extremely small and can be reasonably neglected if the process is not carried out in high-temperature thermal furnaces. As it was shown above, under normal physical conditions, this coefficient is 2.0525 accurate to the fourth digit. In gas analyzers with integrated resistance, to compensate for the influence of temperature, this coefficient is programmed with different, but rather high, accuracy from 2.05 [26] to 2.053 [40]. To recalculate the measured values by gas analyzers of “direct” action, i. e. with an indication of emissions in [ppm], with an excess of oxygen  $K_{V_{\text{O}_2}} = 3\%$  and for normal physical conditions, a coefficient of 2.056 is proposed [5]. Moreover, for normal technical conditions (20 °C and 101325 Pa), the volume in liters of 1 mole of the analyzed gas is more than 7 % different from the value for normal physical conditions, i. e. with an increase in temperature from 0 to 20 °C, the volume of 1 mole will increase from 22.4140 to 24.0551 liters. Then, according to (13), the conversion factor for  $\text{NO}_2$  will decrease from 2.0525 to 1.9125.

The above mechanisms of the formation of pollutant emissions and methodological aspects of determining their magnitude relate exclusively to burner devices. However, important factors affecting the concentration of pollutants in flue gases, primarily nitrogen oxides, are the gas-dynamic and geometric characteristics of the combustion chamber, as well as the conditions for the exit of gases into the chimneys. To determine these dependencies it is necessary to consider several external aspects: the influence of the types of the combustion chamber and their geometry; thermal volumetric load; aerodynamic resistance. Thus, the calculated values of pollutant emissions in flue gases cannot be uniquely determined by the emission class of burner devices. For this, it is necessary to consider the combined system “fuel + burner + combustion chamber + chimney”.

## CONCLUSIONS

1. The mechanisms of formation of pollutants during the combustion of natural gas are considered. It was shown that the formation of NO in boiler furnaces is most significantly affected by the Zeldovich thermal mechanism. The most effective methods and engineering solutions for the design of low emission class burners have been determined.

2. A methodology has been developed for calculating nitrogen oxide emissions depending on the measurement conditions, the emission class of the burners, and the composition of the gas burned. Coefficients are obtained for converting the values of pollutant emissions from specific energy units (generated thermal energy) into specific volume units (flue gas volume).

3. The emission classes of burner devices are determined that correspond to current environmental standards, depending on the capacity of boiler plants.

4. The necessity of taking into account the design of the combustion chamber when calculating the emission of pollutants is shown.

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