Simulation of Nitrogen Oxides Formation as Air Pollution on the Example of Real Combustion Furnace

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Abstract: - As in other countries in Central and West Asia, most of Kazakhstan's power plants have reached their potential as they were built mainly during the Soviet era. In this regard, it is important today to optimize coal consumption processes, introduce new environmentally friendly technologies and reduce emissions. And it is impossible without conducting detailed studies of the combustion of fuels in the combustion chambers of energy devices. It can provide complete information about the nature of complex processes of heat and mass transfer without special financial, time and labor costs. The existing models of the formation and destruction of nitrogen-containing substances NOx do not have a universal nature of use, since the fuel and combustion conditions are different. In this regard, the proposed work considered two models that are widely used. The aim of this work is to select a model that actually can describe the processes of dioxin formation in the combustion chamber of a real CHP boiler when high ash content coal is burned in it. The results of the simulation were verified with the data of field experiments, which allows an objective conclusion to be made. The obtained research data and the method of applying the selected optimal kinetic scheme of dioxin formation contribute to the development of new and optimization of existing measures to control environmental pollution.

Key-Words: - Air pollution, coal combustion, kinetic scheme, nitrogen oxide, simulation, 3D visualisation

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1 Introduction

The global crisis in connection with the COVID-19 pandemic has serious consequences not only for the global economy; it also had an extraordinary impact on various sectors of the energy system of all countries [1]. The results of the impact of the pandemic on all global energy resources are shown in Fig.1.

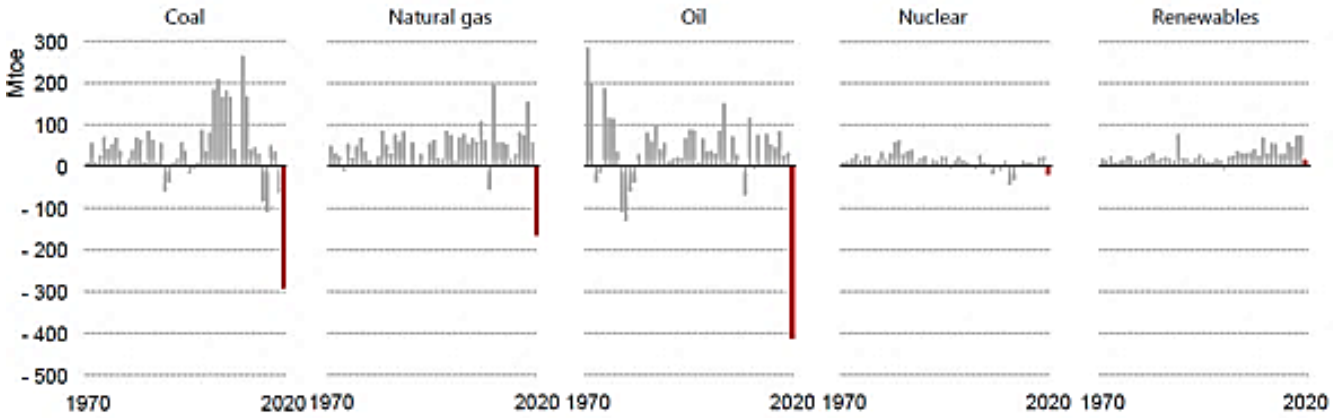


Fig.1. Change in demand for world energy resources by type, 1970-2020 [1]

Despite the fact that coal production declined by 3% in 2019, coal will remain the leading fuel for power generation in the world. A significant increase in coal production will be observed in Asia, especially

in China and Southeast Asia. It's connected with offsetting decline in coal production in advanced economies. For example, the United States of America is expanding its gas production, while Europe is seeing renewable energy generation and coal-to-gas switching. Also, many countries announce the end of the use of coal: for example, the largest consumer of coal in Europe - Germany, plans to get rid of coal by 2038 [2]. Energy is the leading sector of the economy in Kazakhstan, as it has large reserves of fossil fuels and reserves of other minerals and metals. Total energy production in Kazakhstan (178 million tons of oil equivalents in 2018) more than doubles its energy demand. In 2018, Kazakhstan became the 9th largest coal producer in the world (108 million tons); ranked 17th in the world for crude oil production (91.9 million tons) and 24th for natural gas (38.7 billion m^3). Kazakhstan is also a major exporter of energy: in 2018 it was the 9th largest exporter of coal, 9th in crude oil and 12th in terms of natural gas. According to IEA forecasts, the demand for primary energy in Kazakhstan will increase at a rate of 1.8% per year until 2030.

Most power plants in Kazakhstan use coal with a high (over 40%) ash content. Using low quality coal (and even untreated coal) to generate electricity allows the country to save on other fossil fuels. According to academician B.K.Aliyarov, there is currently very little experience in the use of boilers operating on such coal throughout the world [3]. The experience gained in this area in Kazakhstan covers a climate in which temperatures range from + 50 ºC to –50 ºC and can be considered at the global level.

Coal will undoubtedly remain the main source of energy in Kazakhstan's energy balance. Climate change, environmental protection, ash disposal problems and hazardous gases (coal-bed methane) are the main problems facing the coal economy.

Global energy trends such as optimal energy consumption, efficient use of energy, research and development of renewable energy sources are a significant environmental component of the country's energy policy. In the future of the energy policy of the Republic of Kazakhstan, it is planned to develop the raw material base, which will be achieved by enriching and improving the quality of coal, its deep processing to obtain liquid fuel and synthetic substances. The high concentration of methane in coal seams also allows it to be used on a large scale.

World Energy Outlook 2019 [4] outlines a cost-effective strategy to modernize, repurpose and recycle existing facilities to cost-effectively reduce their coal-related emissions. Improving the flexibility and efficiency of the coal-fired power plant fleet is a priority, with R&D funding essential to improve the overall performance, reliability and flexibility of existing coal-fired power plants.

Today in the energy sector of the Republic of Kazakhstan there is an urgent need to optimize coal consumption processes, introduce new environmentally friendly technologies and reduce emissions. As in other countries in Central and West Asia, most of Kazakhstan's power plants have reached their potential as they were built mainly during the Soviet era. Currently, only 14 410 MW of the country's installed capacity is in operation, amounting to about 18 602 MW. To meet the expected 2.5% annual growth in demand, older generation units will need to be replaced and additional units introduced. Efficiency gains through retrofitting existing power plants can significantly reduce CO_2 emissions, can also reduce sulfur dioxide SO_2 and, in some cases, nitrogen oxides (NOx) emissions [5-9].

Improving the efficiency of the energy industry while maintaining its' economic benefits and environmental friendliness during operation is currently based on measures for coal enrichment, as well as optimization of combustion processes. At the same time, an important role is played by research using computational technologies, which contributes to the development of design solutions or the determination of a strategy for modernizing a power facility, as well as its components [10-11].

In this regard, it is important to conduct detailed studies of the combustion of fuels in the combustion chambers of energy devices, which can provide complete information about the nature of complex processes of heat and mass transfer without huge financial, time and labor costs.

The statement of the problem of the processes of fuel combustion in the combustion chambers of an energy facility is accompanied by a number of questions related to the representation of the real conditions of fuel combustion, also with the accuracy and adequacy of the expected results. In this regard, the organization of the formulation of a physical, mathematical and chemical model of fuel combustion in a combustion chamber is of particular importance. The purpose of this work is to determine an effective NOx model suitable for calculating the formation of nitrogen oxides during the combustion of coal fuel with high ash content in an industrial boiler of a Kazakhstan CHP.

2 Nitrogen oxides (NO) formation during fuel combustion

Nitrogenous compounds (NOx, in the sum of NO, NO_2, N_2O, N_2O_2, N_2O_3, N_2O_4 and N_2O_5), which are one of the main pollutants in fuel combustion, are formed from molecular nitrogen N_2 carried with the air and organic nitrogen bound in the fuel (fuel -N). Huge amount of elementary gas-phase reactions are involved in the formation of NOx, which can be formed or destroyed by the listed reaction processes in the gas phase: thermal NOx, fast NOx, fuel NOx and nitrous oxide N_2O. The detailed chemical kinetic mechanisms of NOx have been widely considered in many literatures [12-13]. However, despite the amount of research, there is still a need to develop an effective NOx model, which requires simplification of the generalized reaction mechanisms in the simulation (CFD) of fuel combustion in practical systems: as boilers, different furnaces, internal combustion engines, etc. [14-16]. This allows to reduce the calculation time and to make this laborious method more attractive for industrial applications.

The formation of NO in a particular combustion system is determined by chemical and physical processes interaction taking place inside the combustion chamber. There are three mechanisms of NO formation are known, which differ chemically: the thermal mechanism of NO, the fast mechanism of NO, and the fuel mechanism of NO. The thermal NO mechanism results from thermal dissociation and subsequent reaction of N_2 and O_2 molecules at comparatively high temperatures at fuel-poor flame zone. This process is described by a set of chemical reactions:

$$N_2 + O \overset{k_1}{\leftrightarrow} NO + N, \quad (1)$$

$$N + O_2 \overset{k_2}{\leftrightarrow} NO + O. \quad (2)$$

This mechanism, known as an extended Zeldovich mechanism, has been extended to more accurately describe the formation of thermal NO under fuel enrichment conditions by including an additional elementary reaction:

$$N + OH \overset{k_3}{\leftrightarrow} NO + H. \quad (3)$$

From these reactions, to approximate the rate of thermal nitrous oxides formation the next expression can be received:

$$\frac{dc_{NO}}{dt} = k_{1f} c_O c_{N_2} + k_{2f} c_N c_{O_2} + k_{3f} c_N c_{OH} - \\ -k_{1b} c_{NO} c_N - k_{2b} c_{NO} c_O - k_{3b} c_{NO} c_H . \quad (4)$$

The reaction rate coefficients k_{1f}, k_{1b}, k_{2f}, k_{2b}, k_{3f}, k_{3b} for direct reactions and corresponding reverse reactions can be expressed in accordance with the Arrhenius law:

$$k = AT^{\beta} \exp(-\frac{E_{\beta}}{RT}), \quad (5)$$

where A – is the pre-exponential factor, β is the order of the temperature dependence of the pre-exponential factor, E_{β} – is the activation energy, T – is the temperature, and R is the universal gas constant. The concentration of oxygen atoms O and free radical OH is calculated from empirical relationships.

The NO fast mechanism is formed much earlier than the NO thermal mechanism. This mechanism occurs only in fuel-rich zone of the flame, and it also includes the prompt formation of HCN (hydrogen cyanide), followed by its oxidation to NO. The following reactions are the most likely starting points for fast NO:

$$\begin{aligned} \text{Fuel} &\rightarrow \text{CH, } CH_2\text{...} \\ CH + N_2 &\rightarrow HCN + N \\ CH_2 + N_2 &\rightarrow HCN + NH \end{aligned} \quad (6)$$

The rate of fast NO formation is calculated:

$$\frac{dc_{NO}}{dt} = k c_{O_2}^{b} c_{N_2} c_{fuel} \exp(-\frac{E}{RT}), \quad (7)$$

where c – is the concentration, k – is the pre-exponential factor, b is the reaction order for O_2 (molecular oxygen), and E – is the activation energy.

Fuel NO (about 80% of all nitrogenous compounds) is formed due to oxidation of nitrogen organically bound with fuel and is released in the area of the flame. Since liquid and gaseous fuels do not have chemically bound fuel nitrogen in their composition, it can be argued that the study of the formation of fuel NO's is of paramount importance in the coal burning industry.

The proposed work is aimed to determine the effective NOx model suitable for calculating the nitrogen oxides concentration during the native coal combustion (with high-ash content) in industrial boiler. As the main mechanisms for the formation and destruction of nitrogen oxides were chosen two models (De Soete and the Mitchell-Tarbell) [17].
De Soete's model for the formation of NOx as a key reaction uses a mechanism where 90% of nitrogen oxides are formed via hydrocyanic acid HCN. In addition, in this model, the role of atomic nitrogen N and oxygen concentration O_2 in the oxidizing environment is important. In the Mitchell-Tarbell model, the formation of nitrogen oxides occurs by the reaction of coal pyrolysis, homogeneous combustion of hydrocarbon compounds, heterogeneous combustion of coke residue, the formation of thermal and fuel NH_3 and terminal nitrogen centers CN (HCN, CN, etc.). Both models are widely used in a number of foreign countries to calculate the formation of nitrogen oxides. We have to choose one optimal model, which will most adequately predict the formation of nitrogenous compounds for our low-grade coal and at the same time will be effective when used in a computer program.

2.1 Problem statement

The combustion chamber of the BKZ75 boiler at the Shakhtinskaya CHP (Shakhtinsk, Republic of Kazakhstan) was chosen as the object under study. As a combustion fuel, the Karaganda raw coal was used with the following composition: C - 33.87%, H_2 - 6.63%, S - 1.92%, N_2 - 2.23%, O_2 - 9.65%, W - 10.60%, A - 35.1%. Heat is Q = 34162 kJ/kg and the dispersion of coal dust equal to R_{90} = 20%.
The CFD code of the FLOREAN computer program (FLow REActioN) was used to carry out computational calculations [18-20]. It takes into account all the real conditions of the process under study: the technical parameters of the boiler, the physical characteristics of the fuel, the mechanism of fuel combustion, the processes of heat and mass transfer, the mathematical formulation of the problem and chemical transformations that occur at high temperatures.
Numerical studies of heat and mass transfer processes were carried out using three-dimensional equations of convective heat and mass transfer taking into account convective and radiative heat transfer, chemical kinetics, and two-phase nature of the medium. All these nonlinear equations include the law of conservation of mass, the law of conservation of momentum (Navier-Stokes equation), the law of conservation of energy and the law of conservation of matter [21-23].

3 Simulation results and discussion

The processes of formation and destruction of NOx in combustion systems mainly depend on the temperature and the fuel/oxygen ratio in the combustion zone [24-25].
As can be seen from Fig.2, the combustion zone throughout the entire volume does not exceed the average temperature of 1400 °C. The two-dimensional graph shows that below the burner belt the temperature gives its maximum, and it is explained due to the presence of high turbulence during vortex mixing of injected gas-air fuel from two opposing pairs of burners. In this area of the burners, the minimum temperature values are observed, which increase in the center, then again weaken in height towards the exit from the combustion chamber.
It is known that temperature affects the degree of fuel burn up, which causes the release of volatiles and the remainder of solid coke. From the graphs in Fig.3a and Fig.3b it can be noted that the maximum formation of coke residue and the release of volatile fuel components during combustion occurs at the height of the chamber Z=2-9 m.

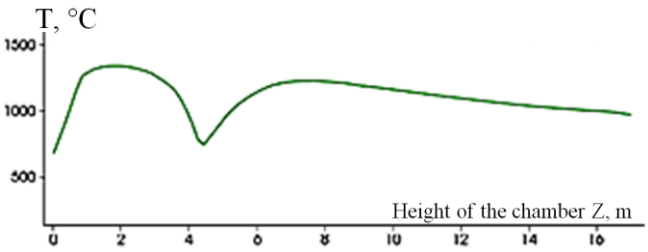
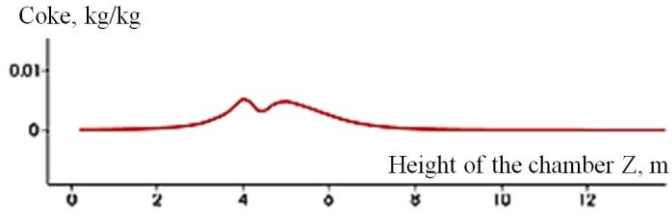


Fig.2. Two-dimensional graph of the temperature distribution along the height of the chamber

a)



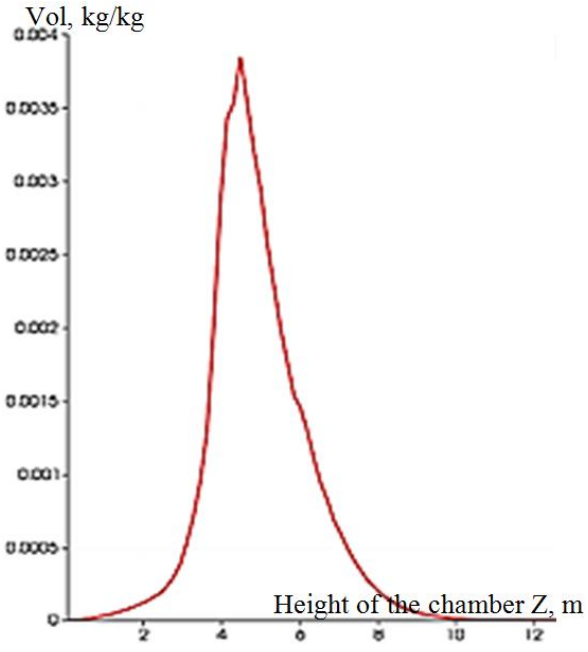


Fig.3. Two-dimensional graphs of the distribution along the height of the chamber for the concentration of a) coke residue and b) volatiles

For Karaganda coal with a high mineral content (ash content over 35%), the degree of burn up along the length of the flame is lower than for other high-quality coals. At the beginning of the flame, this difference is 10%, and by the exit from the combustion space it decreases to 2-3%.

At high temperatures of the combustion zone, more volatile substances are released, and, accordingly, more nitrogen is released, which in a relatively fuel-rich environment are less converted to NO. At lower temperatures, less volatile are produced, which reduces the release of nitrogen from the coal. However, such fuel nitrogen is released in a relatively fuel-poor environment, and this increases the proportion of NO. Table 1 below shows the results of numerical calculations of temperature, coke residue and the yield of volatiles, for two selected sections of the combustion chamber.

Table 1. Results of the computational experiment

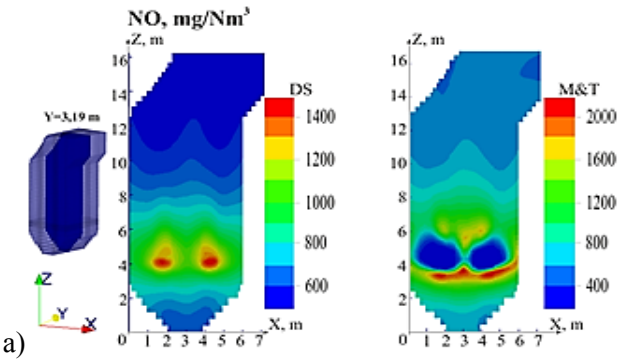
	Temperature, °C	Coke	Volatile
	At the Y=3.19 m Deep in the center		
Max	1339.98	0.0225	0.0104
Mean	1076.13	0.0098	0.0017
Min	534.49	0.0023	$3.43*10^{-8}$
	At the exit section (X=7.04 m)		
Max	978.40	0.0030	$8.16*10^{-8}$
Mean	881.30	0.0020	$1.94*10^{-8}$
Min	706.98	0.0017	$1.41*10^{-9}$

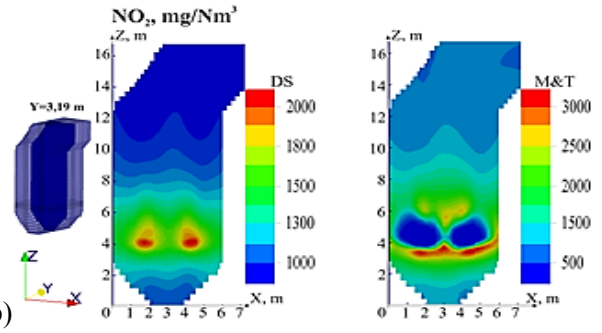
The given average values for the coke residue during the combustion of high-ash coal show that by the exit from the combustion chamber the amount of solid residue is reduced by 80%. This proves that coals with high ash content do not burn out completely, thus fuel NO react less. This requires a special study of the kinetic mechanism of NO formation when burning Kazakhstan coals with high ash content.

As a result of this approach, kinetic schemes for the formation and destruction of NO were selected in this work for studying the formation of nitrogenous substances according to two models using a computer program for three-dimensional modeling. Below are presented results of computational experiments on the formation of nitrogen oxides NO during combustion of high-ash coal in the combustion chamber of the BKZ-75 boiler placed at Shakhtinsk CHP.

The distribution of the concentration fields of nitrogen oxide NO and nitrogen dioxide NO_2 in three-dimensional Fig.4a and Fig.4b, Fig.5a and Fig.5b shows that the formation of NO according to two models (the Mitchell-Tarbell (MT) model and the De Soete (DS) model) differ, although there is a qualitative similarity expressed.

Fig.4a and Fig.4b shows the depth distributions of nitrogen oxide NO and nitrogen dioxide NO_2 in the center of the combustion chamber (section Y = 3.19 m). From the analysis, it can be noted that, according to the De Soete model, the maximum concentrations of nitrogenous substances are observed in the area of burners (for NO above 1400 mg/Nm^3, for NO_2 above 2000 mg/Nm^3), while according to the Mitchell-Tarbell model, the maximum values (for NO above 2000 mg/Nm^3, for NO_2 above 3000 mg/Nm^3) is located below the burner tier and the areas of minima (for NO below 400 mg/Nm^3, for NO_2 below 500 mg/Nm^3) are clearly visible in the belt of burners, which, according to the De Soete model appear near the turning zone of the camera (for NO below 600 mg/Nm^3, for NO_2 below 1000 mg/Nm^3).

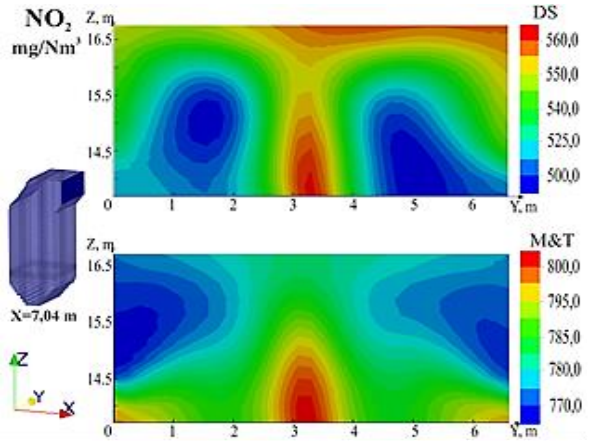




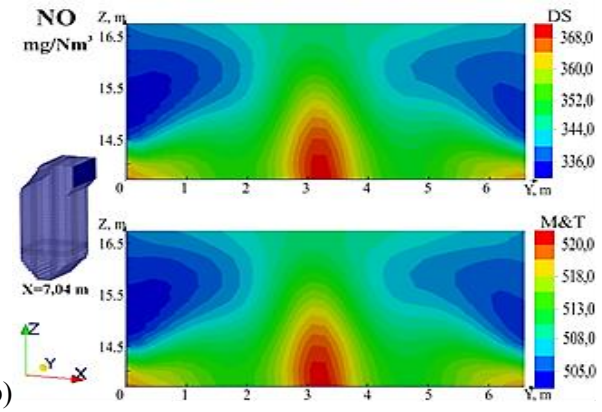
b)

Fig.4. Three-dimensional distributions of a) NO and b) NO_2 concentrations in the section Y=3.19 m of the combustion chamber according to models: De Soete (DS) and Mitchell-Tarbell (M&T)

Analyzing the quantitative characteristics of the concentration fields, it can be seen that higher quantitative values of the NOx concentration are present in the distributions according to the Mitchell-Tarbell (MT) model.
Fig.5a and Fig.5b shows three-dimensional fields of nitrogen oxides NOx concentration at the outlet section of the combustion chamber (X=7.04 m) according to two kinetic models of NOx formation.



a)



b)

Fig.5. Three-dimensional fields of concentration of a) NO and b) NO_2 concentrations in the outlet section of the combustion chamber (X=7.04 m) according to two kinetic models of NOx formation

A decrease in the concentration of nitrogen oxides at the exit from the combustion space is due to the fact that chemical interactions between the components weaken towards the exit.
The qualitative picture of the distribution of nitrogen dioxide concentration differs significantly compared to the distribution of nitrogen oxides according to the De Soete model, while it differs slightly according to the Mitchell-Tarbell model. However, the quantitative indicators of discrepancies are the same and equal to 35%. According to the De Soete model, increased concentrations of nitrogen dioxide are observed in the upper part of the outlet section, this is explained by the fact that the increased temperatures in this area (Fig.6) contribute to the formation of nitrogen dioxides, which did not have time to fully decompose to nitrogen monoxides. Secondly, in this region, an oxygen concentration of 7.3% is observed, which certainly affects chemical interactions.

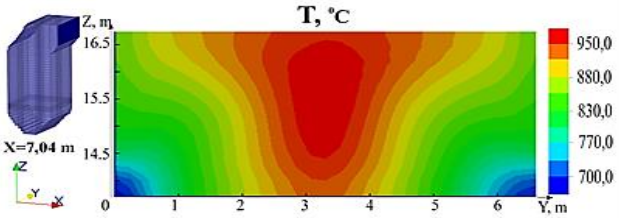


Fig.6. Temperature distribution in the outlet section (X=7.04 m) of the combustion chamber

Below represented the maximum, minimum and mean concentrations of nitrogen oxides NO and oxygen in the sections of the combustion chamber according to models: Mitchell-Tarbell and De Soete (see Table 2).

Table 2. Values of the concentration of substances by models

	DS			MT		
	NO_2	NO	O_2	NO_2	NO	O_2
	At the exit from furnace					
Max	568.36	370.70	0.1132	803.93	524.35	0.1159
Mean	533.97	348.27	0.0732	782.05	510.07	0.0706
Min	504.24	328.88	0.0550	771.18	502.99	0.0545

	At the Y=3.19 m deep in the center					
Max	3335.4	2175.4	0.174	2230.7	1454.9	0.1712
Mean	970.67	633.10	0.085	1258.9	821.12	0.0840
Min	47.47	30.97	0.031	781.74	509.87	0.0311

Analyzing obtained results, it can be seen the difference in the values of NOx and O_2 concentrations according to the selected models for the indicated cross sections is significant and amounts to more than ~40%. The discrepancy between the oxygen concentration values for the two models inside the chamber is less significant (~1.2%), and at the exit from the chamber they are more noticeable and equal to 3.5%.

Fig.7 shows the distribution curves of the average values of the concentrations of nitrogen oxides NOx over the height of the combustion chamber of the BKZ-75 boiler, calculated using the Mitchell-Tarbell and De Soete models. It can be seen that the average concentration values of nitrogen oxides NO according to the Mitchell-Tarbell model exceed the results obtained according to the De Soete model by a factor of 1.65. It also shows the values of the concentrations of nitrogen oxides NO, obtained directly at the TPP [26-27], the MPC value for coal-fired TPPs of the Republic of Kazakhstan and the norms of nitrogen oxide emissions for TPPs of the Republic of Kazakhstan with liquid bottom ash removal.

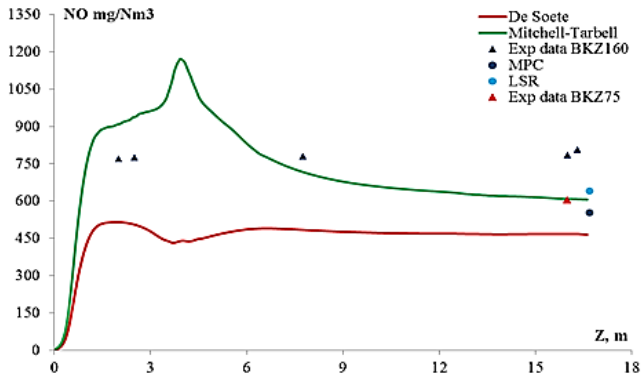


Fig.7. Verification of the results of a computational experiment

The analysis shows that the experimental points and the MPC value are closer to the values obtained by the Mitchell-Tarbell model. At the outlet, the average value of the concentration of nitrogen oxides NO for the first case (Mitchell-Tarbell model) is 613 mg/Nm^3, while for the second case (De Soete model) the value is clearly underestimated.

Such discrepancies in NO concentrations when carrying out computational experiments for the two indicated models is due to the fact that, with the chosen design of the combustion chamber and combustion of high-ash Kazakh coal in it, it provides high-temperature flue gases (more than 1300 °C) and stable ignition of the dust-air mixture, which affect the final result of the calculations. As it turned out, the De Soete model is not sensitive enough to temperature changes in the area of the combustion chamber of the BKZ-75 boiler when high-ash coal is burned in it.

Summing up the work, it can be argued that when conducting computational experiments to study the formation and decomposition of nitrogen oxides NOx when burning Kazakhstani coals with high ash content (30-50%), it is optimal to use the Mitchell-Tarbell model (MT), which reflects real quantitative and qualitative indicators. This choice is taking into account the fact that MT model mainly consider the influence of the mineral part of fuel composition, which will certainly have an effect during the pyrolysis of high-ash coal fuel. Also, this model allows you to make the necessary correlations at high temperatures in the combustion chamber. Due to these advantages, we can firmly speak about the effectiveness of this model when conducting research on the combustion of Kazakhstani high-ash fuels.

The MPC values given during verification in operation are considered acceptable in Kazakhstan; however, they are almost an order of magnitude higher than the values prescribed by the EU Directive. It is also known that the higher the boiler capacity, the greater the formation of nitrogen oxides NOx in the combustion products, which in turn leads to a higher concentration of emissions. Therefore, it is important to conduct further studies to reduce harmful NOx emissions, which include various activities related to the development of design solutions, modernization of the power facility and optimization of combustion processes. Such studies require more mandatory participation of computational technologies and modeling. In this regard, the conclusions drawn in this work can also be useful in solving certain problems.

4 Conclusion

In this work, we have carried out computational studies of the burning of our domestic coal, which has A ~ 40% in the furnace of a real power facility of the Kazakhstan Republic. Analyzing the results obtained and verifying them with field data and maximum permissible values, an optimal kinetic

model was determined for use in computational experiments, which reflects the real picture of the processes of formation of nitrogen-containing substances during coal combustion.

In conclusion, it can be concluded that when carrying out studies of mass transfer processes, namely the formation of nitrogen oxides NO, when burning Kazakhstani high-ash coals in industrial boilers of low productivity, it is preferable to use the kinetic model of Mitchell-Tarbell. The used method of computational experiment makes it possible to obtain adequate data on the processes of formation of harmful dust and gas emissions, particularly, the nitrogenous substances during the combustion of low-grade coal.

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Contribution of individual authors to the creation of a scientific article (ghostwriting policy)

Aliya Askarova, Saltanat Bolegenova has organized and executed the experiments.
Valeriy Maximov and Meruyert Beketayeva carried out the simulation results, interpretation (discussion) and verification of results.

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