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RESEARCH AND ASSESSMENT OF THE ENVIRONMENTAL SECURITY OF BAKERY ENTERPRISES

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Abstract. The paper considers how to decrease the concentration of nitrogen oxides in gas emissions of bakery enterprises. Nitrogen oxides (NO_x) formed in the course of burning natural gas are responsible for more than 90% of an enterprise's hazard category. So, it has been determined how much NO_x is contained in exhaust gases emitted at different loadings on the floor of the oven at an enterprise, this parameter being the main characteristic of the effectiveness of ovens and their impact on the environment. The paper presents the results of experiments that have allowed determining the regression equations describing how the NO_x quantity per unit of output in an enterprise's exhaust gases depends on the amount of the fuel consumed and on the loading on the floor of baking ovens. The procedure of the experiments has been described, and the numerical results have been presented and analysed. It has been established that when making a loaf of wheat-flour oven-bottom bread weighing 0.9 kg, with 70% loading on the floor of an oven (PPC1381), the nitrogen oxides concentration in combustion products is 212.00 μg/m^3, and the specific NO_x formation is 292.25 μg/kg. The nitrogen oxides concentration is 152.00 μg/m^3, and the specific formation is 306.00 μg/kg when the oven floor loading is 40%. If a similar range of products is baked in ovens Minel100 with the maximum-loaded and half-loaded oven floor, the production is accompanied by specific nitrogen oxide emissions of 239.50 μg/kg and 247.80 μg/kg respectively. When enriched buns of 0.1 kg are baked in ovens BN50 with the maximum-loaded and half-loaded oven floor, the process is accompanied by specific nitrogen oxide emissions of 209.20 μg/kg and 265.96 μg/kg respectively. The nitrogen oxides content in gases withdrawn from bakery ovens has been instrumentally measured. This has allowed obtaining regression equations of dependence of the specific NO_x weight in gas emissions on the amount of fuel consumed and on the oven floor loading. It has been found that in the ovens considered, the nitrogen oxide formation per output unit decreased when the oven floor loading increased, because less fuel is needed to make up for the loss of heat accompanying the emission of fumes. The contributions of the argument parameters have been determined according to the regression model to estimate the quantitative dependence. The amount of nitrogen oxides depends on the oven floor loading. A mathematical model has been developed describing how nitrogen oxide formation depends on the oven floor loading and fuel consumption. The model can be used to introduce an industry standard of quantification of nitrogen oxides formed when manufacturing a unit of output.

Keywords: nitrogen oxides, breadbaking industry, exhaust gases, environment.

Introduction. Formulation of the problem

In recent years, it has become a topical problem of how to save resources and reduce the harmful effects of production on the environment [1]. The greatest hazard for the environment and people is nitrogen oxides resulting from industrial activities [2]. For the ozone layer, nitrogen oxides are dangerous because that under the influence of the Sun's soft ultraviolet radiation, which is almost not retained in the stratosphere, nitrogen (IV) oxide decomposes with the release of nitrogen (II) oxide, and the latter is oxidised by ozone [3-4]. As a result of a series of consecutive reactions, one nitrogen oxide molecule is enough to destroy 10 ozone molecules on average [5].

Analysis of recent research and publications

Protecting the atmosphere from pollutants formed in the course of burning any fuel is one of the most serious environmental challenges of our time. Except for water vapour, all other combustion residues adversely affect the environment [6-7].

Nitrogen oxides are not only known to be the most dangerous of all combustion products polluting the atmosphere with toxic substances. They also take an active part in a number of unwelcome processes in various parts of the atmosphere, such as photochemical smog, increased acidity of precipitation, formation of tropospheric and reduction of stratospheric ozone [8-11]. It should be emphasised that in some of these processes, nitrogen oxides behave as catalysts, which makes them particularly dangerous pollutants [12]. Under natural conditions, nitrogen oxides are formed in an amount of about 700 million tons/year as a result of volcanic eruptions, wild fires, air discharges, lightning, as well as in the soil and in the surface layers of the ocean due to anaerobic processes [13].

Industrial plants generate energy by burning various fuel types, and thus, contribute much to anthropogenic nitrogen oxide emission into the atmosphere [14,15]. Mainly, nitrogen oxides are formed during organic fuel combustion at high temperatures (more than 1000°C) and then transformed in the atmosphere into NO_2 [16].

Some scientists [17] believe that in the course of fuel combustion in boilers and other devices, only nitrogen oxides are formed, but on leaving a chimney, they are quickly oxidised to nitrogen (IV) oxide. Other researchers [18] consider that only 40-80% of nitrogen (II) oxide contained in flue gases is converted into nitrogen (IV) oxide on leaving a chimney.

Standards of nitrogen oxide levels vary with each individual country. Thus, in the former USSR, in the 1990s, the one-time MPC was 0.085mg/m^3, and in Japan, the standard was tighter: 0.035mg/m^3 [19]. In developed capitalist countries, they did not take into account that harmful effects were intensified when SO_2 and NO_2 were both present in the air. But the studies by some foreign and domestic researchers [20] have shown that the combination of these oxides at concentrations equal to the sum of the MPC increases their harmful effects on living organisms (cumulative effect).

A significant amount of nitrogen oxides is produced by thermal power plants, metal industries, heating plants, and motor vehicles [21-23].

There are three ways of nitrogen oxide formation, which differ in origin, but have the same chemical composition:

– thermal nitrogen oxides (thermal NO_x)

– prompt nitrogen oxides (prompt NO_x);

– fuel nitrogen oxides (fuel NO_x) [24].

The so-called thermal nitrogen oxides are formed at high temperatures (T>1500 K) and with high oxygen concentrations during the oxidation of atmospheric nitrogen in the course of combustion.

Prompt nitrogen oxides are formed when nitrogen from the air is fixed with hydrocarbon radicals during combustion of fuel. In this way, oxides are formed at a very high rate (hence their name *prompt*). With fuel mixtures used and with low-temperature combustion, they can make up as much as 25% of the total nitrogen oxides content.

Fuel nitrogen oxides are formed during oxidation of nitrogen-containing substances in the fuel. Concentrations of fuel oxides can be quite significant if nitrogen-containing substances present in the fuel make up more than 0.1% of its weight. As a rule, this only applies to liquid and solid fuels [25].

In the European Union, EU Directive 2008/50/EC (21 May 2008) obligates all industrial enterprises to bring their emissions of harmful substances into the atmosphere, including NO_x, in accordance with the standards established. In 2011, Ukraine ratified EU Directive 2001.80 on limiting hazardous emissions from enterprises and bringing its emission standards to the European ones. To date, a number of technological measures have been developed and largely implemented to reduce the NO_x content in exhaust gases [26,27]. However, the effectiveness of these measures is clearly insufficient.

In this regard, improving the existing technologies and developing new ones to reduce the NO_x content in exhaust gases are tasks that are becoming more and more important.

Nitrogen oxides formed during natural gas burning account for over 90% of the hazard category of bakery enterprises. That is why the main parameter characterising the efficiency of ovens and their effect on the environment is the NO_x content of exhaust gases [28,29].

The purpose of this research is to determine how at bakery enterprises, NO_x emissions in exhaust gases correlate with different loading on the oven floor, and how they can be minimised. **The objectives** of the study are as follows:

1. studying how nitrogen oxides affect the environment;
2. characterising the ways of nitrogen oxide formation at enterprises;
3. substantiating, using a mathematical model, the system of actions to prevent bakery enterprises from polluting the environment.

Research materials and methods

The control samples of exhaust gases were taken at the bakery enterprise *TOV Odesky Khlib* in Odessa.

Before the experimental studies, the fuel consumption in the oven burners was checked and regulated. To this end, the fuel pressure before the burners was equalised in accordance with the readings of the standard pressure gauges. The optimal values of air surplus on exit from the oven were determined without violating the norms of the technological regulations for oven operation. With each loading mode, 5 rough tests and 2 main (balance) tests were performed. To determine the content of nitrogen oxide NO and nitrogen dioxide NO_2, a portable gas analyser PEM-4M2 was used. Air samples were taken and analysed in accordance with [30,31]. Taking each sample consisted in a succession of 3 samplings at a particular sampling point. The duration of sampling was the same for each sample, being no less than 20 minutes.

All calculations were performed on a PC using Excel, in particular, the REGRESSION software module. The calculation results are summarised in Table 1. The accuracy and quality of the regression model have been estimated by the following parameters: residual variance coefficient S_{res}, coefficient of determination R^2, average relative error E%. The overall quality of the regression equation and its statistical reliability were estimated using the F-test (Fisher statistics). To this end, the value F_n was calculated according to the data in the model, the reference value (table value) F_{tabl} was selected, and F_n was compared with F_{tabl}. The first-order autocorrelation in the residuals was determined using the Durbin–Watson statistic (d_p) with two bounds – d_1 and d_2, where d_1 is the lower bound, d_2 is the upper one. The r_{x1x2}, r_{yx1}, r_{yx2} coefficient (pair correlation coefficient) was used to determine the degree of density of the relationship between the two variables. The level of the factor's influence on the function was estimated using the coefficient of elasticity and the beta coefficient (β_{X1}, β_{X2}) for each argument of the model.

Results of the research and their discussion

Formation of nitrogen oxides during fuel combustion is described in the works by domestic and foreign researchers [19,29]. Computational and analytical models of the fuel combustion process and formation of nitrogen oxides have been developed. The problem of minimising nitrogen oxide emissions into the environment is considered in [28]. However, the existing mathematical models do not take into account the different loading of industrial ovens.

Fig. 1 shows how different loadings on the oven floor change the nitrogen oxide and nitrogen dioxide NO_x concentrations in emissions from an oven PPC-1381 when baking wheat flour oven-bottom bread weighing 0.9 kg. The nitrogen oxides content in the flue gases was measured with a gas analyser PEM-4M.

The results of instrumental measurement of the nitrogen oxide content in exhaust gases from industrial baking ovens PPC-1381 have allowed obtaining a regression equation describing how NO_x produced depends on the amount of fuel consumed and the oven floor loading:

$$Y = 321.213 - 4.539 X_1 + 174.671 X_2 \qquad (1)$$

where X_1 – ratio of the quantity of products obtained to the area of the oven floor, kg/m^2,

X_2 – gas volume, m³.

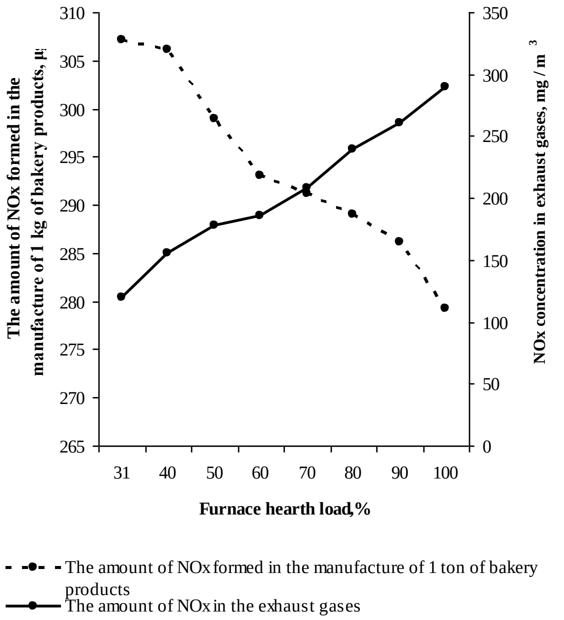

Fig. 1. Determining the nitrogen oxides content in emissions from an oven PPC 1381 when baking wheat flour oven-bottom bread weighing 0.9 kg, with different loadings on the oven floor

The contributions of the argument parameters have been determined according to the regression model to estimate the quantitative dependence. According to the calculations in Table 1, the amount of nitrogen oxides formed when baking 1 kg of bread almost completely depends on the oven floor loading.

The data in Table 1 show that the resulting model has a high coefficient of determination. It is not collinear and can be used to optimise the content of nitrogen oxides in exhaust gases of baking ovens PPC 1381.

Fig. 2 shows how different loadings on the oven floor change the nitrogen oxide concentrations in emissions from an oven Minel 100 when baking wheat flour pan-loaf bread weighing 0.9 kg. The plot is based on the measurements with a gas analyser PEM-4M.

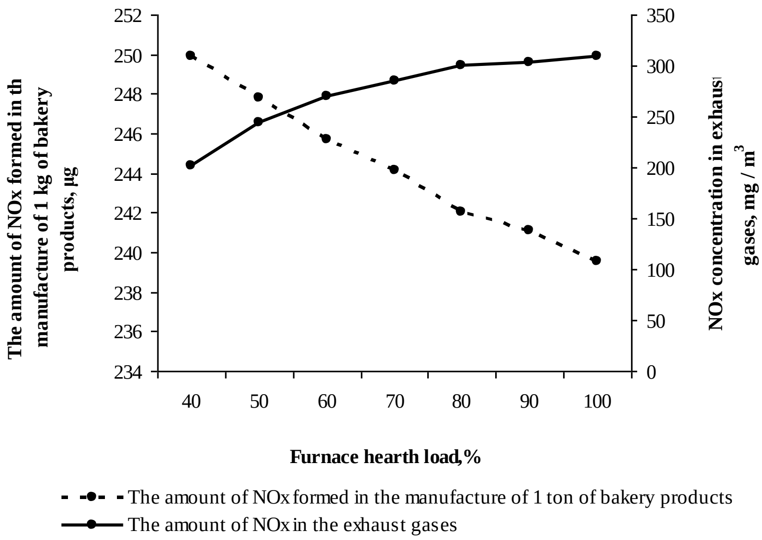

Fig. 2. Determining the nitrogen oxides content in emissions from an oven Minel 100 when baking wheat flour oven-bottom bread weighing 0.9kg, with different loadings on the oven floor

The results of instrumental measurement of the nitrogen oxide content in exhaust gases from industrial baking ovens Minel 100 have allowed obtaining a regression equation describing how NO_x produced depends on the amount of fuel consumed and the oven floor loading:

$$Y = 246.003 - 0.548X_1 + 64.962X_2 \quad (2)$$

where X_1 – ratio of the quantity of products obtained to the area of the oven floor, kg/m^2,

X_2 – gas volume, m^3.

The contributions of the argument parameters have been determined according to the regression model to estimate the quantitative dependence. According to the calculations in Table 1, the amount of nitrogen oxides formed when baking 1 kg of bread almost completely depends on the oven floor loading.

The data in Table 1 show that the resulting model has a high coefficient of determination, and its absolute and relative errors are but insignificant. So, it can be used to optimise the content of nitrogen oxides in exhaust gases of baking ovens Minel 100.

Fig. 3 shows how different loadings on the oven floor change the nitrogen oxide concentrations in emissions from an oven BN 50 when baking enriched buns weighing 0.1kg. The plot is based on the measurements with a gas analyser PEM-4M.

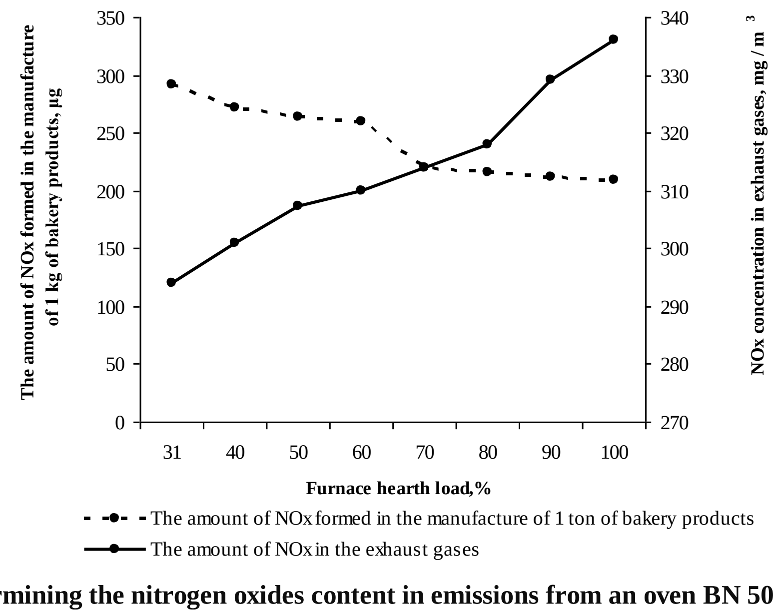

Fig. 3. Determining the nitrogen oxides content in emissions from an oven BN 50 when baking enriched buns weighing 0.1 kg, with different loading on the oven floor

The results of instrumental measurement of the nitrogen oxide content in exhaust gases from industrial baking ovens BN 50 have allowed obtaining a regression equation describing how NO_x produced depends on the amount of fuel consumed and the oven floor loading:

$$Y = 310.847 - 3.349X_1 + 32.704X_2 \qquad (3)$$

where X_1 – ratio of the quantity of products obtained to the area of the oven floor, kg/m^2,

X_2 – gas volume, m^3.

The contributions of the argument parameters have been determined according to the regression model to estimate the quantitative dependence. According to the calculations in Table 1, the amount of nitrogen oxides formed when baking 1 kg of bread almost completely depends on the oven floor loading.

The data in Table 1 show that the resulting model has a high coefficient of determination, and its absolute and relative errors are but insignificant. So, it can be used to optimise the content of nitrogen oxides in exhaust gases of baking ovens BN 50.

All calculations were performed on a PC using Excel, in particular, the REGRESSION software module. The calculation results are summarised in Table 1.

All multiple regression models are statistically significant, and accurate enough. The values of E% never exceed 10%. The coefficients of determination R^2 exceed 0.99, that is, unknown factors account for less than 1%. The signs before the regression coefficients at the arguments correspond to the economic sense of the influence of the arguments on the function: when factor X_1 increases by one, this leads to a decrease in Y by the regression coefficient value, and on the contrary, when factor X_2 increases by one, the function Y, too, increases by the regression coefficient value for this argument.

Verification of how the preconditions of the method of least squares are fulfilled has confirmed the absence of heteroscedasticity and autocorrelation in the residuals.

A comparison of the β-coefficients by their modulus for the model's arguments shows that the influence of factor X_1 on the function is greater. A similar conclusion can be drawn about the coefficients of elasticity.

With factor X_1 having more influence on the function, models of simple regression have been additionally constructed with this argument. The accuracy of the models is slightly lower, but all models are statistically reliable, and the MLS preconditions for them are fulfilled.

The experimental studies have revealed a decrease in the specific formation of nitrogen oxides in the ovens under study when the oven floor loading increased. Thus, with 70% loading of ovens PPC 1381, when baking wheat-flour oven-bottom bread weighing 0.9kg, the concentration of nitrogen oxides in the combustion products is 212.00μg/m^3, and the specific NO_x formation is 292.25μg/kg. The nitrogen oxides concentration 152.00μg/m^3 and the specific formation 306.00μg/kg are observed at 40% loading of the oven floor. If a similar range of products is baked in ovens Minel 100 with the maximum-loaded and half-loaded oven floor, the production is accompanied by specific nitrogen oxide emissions of 239.50μg/kg and 247.80μg/kg respectively. Baking enriched buns of 0.1kg in ovens BN 50 with the maximum-loaded and half-loaded oven floor is accompanied by specific nitrogen oxide emissions of 209.20μg/kg and 265.96μg/kg respectively.

Table 1 – Results of the calculations

Indicates	Oven type					
	PPC 1381		Minel 100		BN 50	
Product type	Wheat-flour oven-bottom bread, 0.9 kg	Wheat-flour oven-bottom bread, 0.9 kg	Wheat-flour oven-bottom bread, 0.9 kg	Wheat-flour oven-bottom bread, 0.9 kg	Enriched buns, 0.1 kg	Enriched buns, 0.1 kg
Regression equation	$Y=321.213-4.539X_1+174.671X_2$	$Y=354.468-6.081X_1$	$Y=246.003-0.548X_1+64.962X_2$	$Y=255.107-0.775X_1$	$Y=310.847-3.349X_1+32.704X_2$	$Y=319.858-3.948X_1$
E,%	0.30	0.377	0.08	0.18	0.26	0.26
R^2	0.992	0.99	0.993	0.972	0.990	0.989
S_{res}	1.57	1.63	0.342	0.618	1.12	1.12
F_p	89.64	82.65	98.4	30.23	74.24	74.7
F_{tabl}	4.88	4.21	6.16	4.95	4.88	4.21
d_p	1.72	2.24	2.39	1.51	1.5	1.2
d_1	0.56	0.76	0.47	0.7	0.56	0.76
d_2	1.78	1.33	1.9	1.4	1.78	1.33
r_{x1x2}	0.604		-0.895		-0.017	
r_{yx1}	0.286	0.381	0.214	-0.54	0.476	-0.31
r_{yx2}	-0.268		-0.214		-0.458	
$\hat{\beta}_{X1}$	-0.742	-0.995	-0.697	-0.986	-0.843	0.994
$\hat{\beta}_{X2}$	0.257		0.32		0.157	
Notes	Two-factor regression	Unifactor regression	Two-factor regression	Unifactor regression	Two-factor regression	Unifactor regression

The data obtained during our research are consistent with the findings of domestic and foreign researchers who studied the problem of how to minimise nitrogen oxide emissions [28]. The decrease in nitrogen oxides emitted resulting from changes in the oven floor loading is confirmed by the conclusions of previous investigations [29].

Thus, to reduce harmful emissions into the atmosphere per product weight unit, an enterprise should increase production volumes and, accordingly, load the ovens as much as possible.

The resulting mathematical models of multiple-factor regression can be used for predictive calculations. It should be expected that the function's prediction accuracy will be quite high. With factor X_1 having a stronger influence on function Y, it is this very factor that can be controlled in production to achieve the predetermined amount of NO_x emissions. Unifactor models can also be used for the purpose.

Approbation of results

The research results obtained have been tested and introduced into production at the Ukrainian bakery enterprise *TOV Odesky Khlib.*

Conclusion

The problem of the impact of nitrogen oxides on the environment has been considered. The research results prove that the main anthropogenic factor of bakery enterprises' environmental impact is the baking process, which is accompanied by thermal and gas emissions.

In the course of the research, methods of resource saving and decreasing the environmental hazards of production have been found. It has been established that an increase in the oven floor loading leads to a decrease in nitrogen oxide formation per unit of output in the ovens under study. This is explained by a decrease in fuel consumption to make for the heat lost with flue gases.

Thus, if the loading in industrial baking ovens PPC 1381, Minel 100, and BN 50 is increased up to 90%, it allows reducing emissions of nitrogen oxides per unit of output by 45–48%. The studies have shown that nitrogen oxide emissions per unit of output are minimum at 80–90% loading of baking ovens.

As a result of the research, a mathematical model has been developed describing how nitrogen oxide formation depends on the oven floor loading and fuel consumption. The model can be used to introduce an industry standard of quantification of nitrogen oxides formed when manufacturing a unit of output.

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ДОСЛІДЖЕННЯ ТА ОЦІНКА ЕКОЛОГІЧНОЇ БЕЗПЕКИ ХЛІБОПЕКАРСЬКИХ ПІДПРИЄМСТВ

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Анотація. Розглянуто можливість зниження концентрації оксидів Нітрогену в газових викидах хлібопекарських підприємств. Утворені в процесі горіння природного газу оксиди Нітрогену складають більше 90% категорії небезпеки підприємства. Тому в якості основного показника, що характеризує ефективність роботи печей та вплив на навколишнє середовище, визначено залежності вмісту в відпрацьованих газах NO_x при різних навантаженнях на під печі на підприємстві. Наведено результати експериментальних досліджень з визначення регресійних рівнянь залежності кількості NO_x на одиницю продукції у відпрацьованих газах підприємства від кількості спожитого палива і завантаження поду хлібопекарських печей. Встановлено, що при завантаженні пода печей ППЦ-1381 на 70% при випіканні хліба подового з пшеничного борошна масою 0.9 кг концентрація оксидів Нітрогену в продуктах горіння становить 212,00 мкг/м, питоме утворення NO_x-292,25 мкг/кг. Концентрація оксидів Нітрогену 152.00мкг/м^3 з питомим утворенням 306,.00мкг/кг відповідає завантаженню пода на 40%. Вироблення аналогічного асортименту на печах Мінел-100 з максимальним і половинним завантаженням пода супроводжується питомими викидами оксидів Нітрогену відповідно 239,50мкг/кг і 24,8мкг/кг. Випічка здобних булочних виробів масою 0.1кг на печах БН-50 з максимальним і половинним завантаженням пода супроводжується питомими викидами оксидів Нітрогену відповідно 209,20 мкг/кг і 265,96мкг/кг. За результатами інструментального визначення вмісту оксидів Нітрогену у відведених газах хлібопекарських печей отримані регресійні рівняння залежності питомої маси NO_x у газових викидах від кількості спожитого палива й завантаження пода. Виявлено зниження утворення оксидів Нітрогену на одиницю продукції в печах, які досліджувались, при підвищенні завантаження пода, що пояснюється зниженням витрати палива на покриття втрат тепла з димовими газами. Отримано математичну модель впливу завантаження пода печі і витрати палива на утворення оксидів Нітрогену, яка може бути використана для впровадження галузевого нормативу з визначення кількісних показників оксидів Нітрогену, що утворюються при виробництві одиниці продукції.

Ключові слова: оксиди Нітрогену, хлібопекарська галузь, відведені гази, навколишнє середовище.

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