ЕКОЛОГІЧНА БЕЗПЕКА, ОХОРОНА ПРАЦІ

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ENVIRONMENTAL ASSESSMENT OF THE INTERMETALLIC CATALYSTS UTILIZATION EFFICIENCY FOR DEACTIVATION OF THE POLLUTANTS EMITTED BY ELECTRODE PRODUCTION ENTERPRISES

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ЕКОЛОГІЧНА ОЦІНКА ЕФЕКТИВНОСТІ ЗАСТОСУВАННЯ ІНТЕРМЕТАЛІДНИХ КАТАЛІЗАТОРІВ ДЛЯ ЗНЕШКОДЖЕННЯ ВИКИДІВ ПІДПРИЄМСТВ ВИРОБНИЦТВА ЕЛЕКТРОДІВ

Purpose. The improvement of the processes of pollution abatement at electrode production enterprises aimed to raise air quality and comply with the environmental standards established in Ukraine.

Methodology. The catalysts efficiency was tested in the stream of exhaust gases produced by kilns and graphitization furnaces in a laboratory at temperatures from 100 to 500 °C and the volumetric gas flow rate W from $30 \cdot 10^3$ to $120 \cdot 10^3$ m³/m³-hr. In order to assess compliance with sanitary and hygienic standards of the area affected by the emissions, the ground-level concentrations of pollutants were calculated using the PLEINAIR-1.25 software before and after the introduction of the catalytic purification of exhaust gases. The software works in accordance with the document OHД-86 "Method of Calculation of the Harmful Substances Concentration in the Air Polluted by Industrial Emissions" and was approved by the Ministry of Ecology and Natural Resources of Ukraine.

Findings. The modeling of the pollutants dispersion in the air showed exceeding ground-level concentrations in the residential area taking into account the initial levels of carbon monoxide, naphthalene, and phenol, which are 1.05 MPC, 1.1 MPC and 1.14 MPC respectively. The results proved the need for measures to minimize the emissions of these substances. We carried out comparative tests using the existing and the newly developed catalysts for neutralization of the exhaust gases from the kilns and graphitization furnaces. The results of the comparative tests showed that the developed catalyst cycle life is 1.6 1.7 times longer than that of the known nickel catalyst.

Application of the newly developed catalyst raised the degree of gases purification to 99.9%. The neutralization of the waste gases vented by the kilns and graphitization furnaces using the developed catalyst resulted in the decrease of the ground-level concentrations of pollutants in the residential areas to the background level.

Originality. We proved theoretically and confirmed experimentally that the catalytic neutralization of carbon monoxide and hydrocarbons by intermetallic catalysts results in the increase of environmental safety of the emissions.

Practical value. The method of deactivation of the waste gases containing carbon monoxide and hydrocarbons emitted from the kilns and graphitization furnaces by means of the intermetallic catalysts has been developed and recommended for implementation at the enterprise PAT "Ukrgrafit".

Keywords: gaseous waste, carbon oxide, hydrocarbons, neutralization, intermetallide catalyst

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Formulation of the problem. Power plants, internal-combustion engines, and industrial plants are the sources of the environment pollution with carbon monoxide (CO) and hydrocarbons (C_mH_n). Chemical and biological properties of CO and C_mH_n and their significant volumes in gaseous emissions increase environmental hazard around the facilities emitting them. Currently, the Ukrainian government introduces standards on emissions to create the basis for the regulation of atmosphere air pollution. The official name of the document is "The Standards of Maximum Permissible Emissions of Polluting Substances from Stationary Sources". It was approved by the Order no. 309 of the Ministry of Ecology and Natural Resources of Ukraine dated June 27, 2006. The concentration of CO and C_mH_n in the emissions produced by most of these sources does not meet the established standards and is a factor of intensive deterioration of the air quality [1]. In order to achieve the established standards, enterprises have to optimize their technological processes, introduce re-equipment and new technologies.

The analysis of recent researches. Electrode manufacturing enterprises vent about 4-5 million cubic meters of gases per hour. Those gases contain CO and resinous substances consisting of a mixture of polycyclic aromatic hydrocarbons, some of which are carcinogenic. It is generally recognized that benzopyrene serves an indicator of the carcinogenic hazard of electrodes production. Its content in the emitted aerosol resinous substances range from 0.8-0.10 to 1-2%. On average, one enterprise vents about 60-80 kg of benzopyrene per day [2].

The high environmental hazard of toxic organic substances determines the importance of the introduction of treatment technologies, which use sorption, thermal and catalytic methods [3–4].

Absorption techniques are widely used in industry for the elimination of various impurities from gases, including CO. However, they are inapplicable in the case of electrodes production process due to the high gas-flow rate, the complex technological scheme of elimination of dust and other contaminants, high pressure and high cost.

The methods of adsorptive gas purification are used mostly for neutralization of vapor-state impurities in the waste gases. These methods have significant drawbacks. For instance, they are material intensive, require large capital expenditures, and, which is the most important, they do not provide the desired degree of gas purification in some cases.

The methods of thermal afterburning of CO to $\rm CO_2$ at high temperatures (800–1200 °C) can be used if the CO concentration is more than 12 %, i. e. in the case when the CO content is above the explosion limit of CO gas mixture. Sometimes the thermal method, even with a high degree of oxidation of organic substances, does not ensures the maximum allowable concentration (MAC) of carbon monoxide in the air. Therefore, in some cases, this method is used only as the first stage of purification.

One of the most effective ways of abatement of CO and C_mH_n is the catalytic method. The catalytic process of neutralization of combustion products runs, as a rule, at temperatures above 300 °C and under short contact times, due to the high flow rate of industrial emissions. The main advantages of the catalytic process in comparison with thermal afterburning consist in its technological and operational characteristics, namely: high efficiency and economy, the absence of harmful side effects.

The tightening of sanitary standards on environmental conditions requires searching for efficient catalysts for neutralization of exhaust gases of industrial plants [4].

For the catalytic elimination of CO and C_mH_n both metal and oxide catalysts can be used. However, upon utilization of the oxide catalysts, the sufficiently high speed of the contact process can be achieved only at relatively high temperatures (300–400 °C). The oxides of some metals may be arranged in the following sequence by the specific catalytic activity value [5]

$$TiO_2 < V_2O_5 < Cr_2O_3 < ZnO < Fe_2O_3 < NiO < CuO < \\ < Co_3O_4.$$

The specific catalytic activity of the mentioned oxides at the temperature of 300 °C differ by more than five orders of magnitude. Oxides of cobalt, copper, nickel and iron have the highest catalytic activity.

Metal catalysts have higher activity in the reaction with CO and C_mH_n than oxide catalysts. The metals may be arranged in the following sequence by the specific catalytic activity at the temperature of 180 °C [6]

$$V < Cr < Mn < Cu < Ag < Au < Fe < Co < Ni < Pd < Pt.$$

Among the basic metals, the highest catalytic efficiency values belong to nickel, cobalt, and iron. What is more, by the specific catalytic efficiency nickel is superior to cobalt by 2.1 times and iron by 16 times. Platinum and palladium have the best properties for catalytic oxidation of CO and C_mH_n . However, platinum and palladium are slightly better than nickel by the specific catalytic efficiency: platinum is 6 times more efficient than nickel, and palladium is 5 times more efficient. On the other hand, nickel is much more active than oxide catalysts. The specific catalytic efficiency of nickel at the temperature of 180 °C is 3 orders of magnitude higher than that of cobalt oxide, and 6 orders of magnitude higher than that of zinc oxide at 300 °C. Thus, as low-temperature catalysts for CO and C_mH_n oxidation, nickel and cobalt catalysts make the most sense. Taking into account the higher activity and lower cost and scarcity of nickel as compared with cobalt, the choice of nickel catalysts for commercial use is the most rational. Thus, among the most effective catalysts are materials containing intermetallic compounds of nickel because of the ability to transfer active oxygen involved in the oxidation process. In this regard, it seems promising to study the properties of the intermetallic compounds of these metals as catalysts.

The objective. The work is intended to improve the processes of neutralization of gas emissions at electrode production enterprises to ensure air quality and meet the environmental standards established in Ukraine. To achieve the goal it is necessary to develop technical solutions for the protection of atmospheric air from the emissions containing CO and C_mH_n , and carry out its field testing at an electrode production enterprise.

Results and discussion. To reduce the emissions of CO and C_mH_n , an intermetallic catalyst has been created. It consists of nickel and aluminum with additions of copper, cobalt and manganese. The compound was produced by self-propagating high-temperature synthesis (SHS). The previously made researches [7, 8] showed several advantages of the porous SHS materials in comparison to similar materials that are synthesized by conventional methods of powder metallurgy. First, we should highlight the high final porosity of SHS materials, which may be higher by 15–20%. Another advantage of SHS materials is their surface area, which is 1.5–1.7 times higher than that of the sintered materials with the same porosity. For example, the specific surface of the SHS-produced catalyst based on the phase NiAl₃ is $108 \text{ m}^2/\text{g}$, [7].

The new intermetallic catalyst passed pilot tests at the company PAT "Ukrgrafit" because the enterprise vents CO (\approx 82 %) and toxic hydrocarbons (\approx 5.5 %), such as benzopyrene, benzene, styrene, and phenol.

The PAT "Ukrgrafit" is a leading Ukrainian manufacturer of graphite electrodes for electric-steelmaking, ore-smelting and other electric kilns; carbon cathode and anode paste for electrodes, carbon paste; lining materials (blocks) on the basis of carbon for the enterprises of the metallurgical, machine-building, chemical and other industries.

The industrial site of the PAT "Ukrgrafit" is a part of the industrial unit that is located to the northeast from the main residential area of Zaporozhye, in its eastern part, on the left bank of the river Dnipro. Air pollution is the main way the PAT "Ukrgrafit" affects the environment and the surrounding residential and public areas.

In the process of baking of extruded electrodes (the so-called "green" electrodes), one section of the kiln vents about 4 thousand m³/h of gas. In the process of baking of impregnated electrodes, it vents 1.5–2.0 times less [2]. In both cases, a sharp increase in the concentration of harmful components in the gas phase (CO, CH₄, resinous substances such as benzopyrene) occurs at the beginning of electrode baking. This is caused by the release of low-boiling hydrocarbons from the coal-tar asphalt, for example, benzene, and by the pyrolysis reaction, when they break apart into simpler units, which goes at relatively low temperatures in the oxidizing atmosphere under the arch of the kiln (natural gas burner).

The main toxic components released into the air from electrodes graphitization furnaces are CO, SO_2 , and resinous substances. The concentration of SO_2 in the gas phase reaches 12-33 mg/m³; the concentration of dust in the gases is 17-25 mg/m³.

From the beginning, the CO content in the gas phase rises progressively as the current capacity increases and reaches a maximum value of 770 mg/m³.

Starting from the 10th—13th hour of operation of the kiln, the temperature of the gases released from the gross mass of carbonaceous materials under the kiln cowl reaches the CO autoignition temperature. Thus, a considerable part of it oxidizes at the higher (upper) levels of filling. Starting from this period, the concentration of CO in the gaseous phase does not correspond to the quantity of CO that is released from the kiln as far as the temperature in the working zone of the kiln continues to increase considerably.

The waste gases contain resinous substances during the whole electrodes graphitization process. Their concentration increases dramatically from 1.5–2.0 to 8–9 mg/m³ during the 37th hour of operation of the furnace when the temperature of the furnace-charge insulation layer rises significantly.

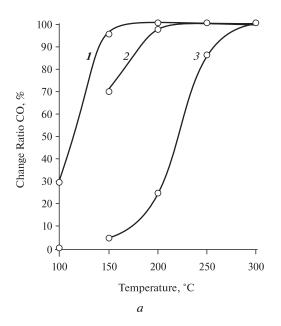
For neutralization of the exhaust gases from the kilns, at the plant of the PAT "Ukrgrafit", they apply electrical purification method. I.e. the flow of the exhaust gases with the rate of $\approx 40~000$ in m³/h is forwarded from the breeching to the C-type electrostatic precipitator, where it is cleaned from the resinous substances and then vented through the flue gas stack to the atmosphere. In the process, the content of CO, SO_2 , and NO_2 in the gas phase does not change, while resinous substances concentration is reduced by 8-90 %. The advantage of this purification method is the implementation simplicity and low cost. However, this method has a significant drawback since it does not allow removing all of the toxic components from the gas (the entire quantity of CO is released to atmosphere).

To date, the waste gases from graphitization furnaces of the PAT "Ukrgrafit" have been emitted to the atmosphere without purification.

The pilot tests of the exhaust gases neutralization by means of the designed intermetallic catalysts were held in the graphitization furnaces of the PAT "Ukrgrafit". We tested the catalyst of known composition (52 % Al + 48 % Ni), the catalyst containing 0.2 %wt. of platinum spread on alumina and the new optimum composition that we have developed, 30 % Ni + \pm 10 % Co + 11 % Mn + 2 % Cu + 47 % Al [9].

The tests of the catalysts activity were conducted on the waste gas flow of the kiln and graphitization furnace by means of the laboratory-scale plant at a temperature from $100 \,^{\circ}\text{C}$ to $500 \,^{\circ}\text{C}$ and volumetric flow rate W ranging from $30 \cdot 10^3$ to $120 \cdot 10^3$ m³/m³ · hr. The concentration of hydrocarbons in the industrial emissions was measured by gas chromatography using the device "Crystal 2000 M". The concentration of carbon monoxide was measured by means of the gas analysis instrument "Palladium-3".

The results of the tests conducted in the flow of exhaust gases showed that the efficiency of the new catalyst proposed and the catalyst containing 0.2 %wt. of platinum differ slightly (Fig. 1, a, b). The highest degree of conversion at a lower temperature was observed



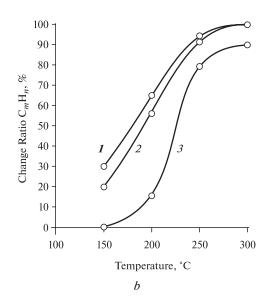


Fig. 1. Catalytic effect of the catalysts under review: a – in the oxidation of CO, %; b – in the oxidation of C_mH_n , %; 1 – Pt-catalyst; 2 – 30 % Ni + 10 % Co + 11 % Mn + 2 % Cu + 47 % Al; 3 – 52 % Al + 48 % Ni

in both cases. In the flow with $W = 30 \cdot 10^3 \, \mathrm{m}^3/\mathrm{m}^3 \cdot \mathrm{hr}$, under the influence of the catalysts the degree of CO neutralization reached 99.9 % at the temperature of 200 °C. The same degree for $C_m H_n$ was reached under 300 °C. The catalyst with the 52 % Al + 48 % Ni composition showed significantly lower activity. The oxidation of CO and $C_m H_n$ was carried out in the flows with different volumetric flow rates. The results showed that with the increase of the volumetric flow rate from $30 \cdot 10^3$ to $120 \cdot 10^3 \, \mathrm{m}^3/\mathrm{m}^3 \cdot \mathrm{hr}$ at the temperature of $300 \, ^\circ\mathrm{C}$ the degree of oxidation of CO and $C_m H_n$ decreases from 99.9 to 95 % in case of utilization of the new catalyst and the catalyst containing 0.2 %wt. of platinum. For the 52 % Al + 48 % Ni catalyst the decrease was from 98 to 75 %.

The catalyst with the composition of 52 % Al + +48 % Ni worked stably over 450 hours, after which the degree of gas purification reduced sharply to 85 %. The new catalyst worked for 720 hours. The degree of purification reduced to 99 %. Because the exhaust gases of the graphitization furnace contain SO₂, the activity of the catalyst with the composition of 52 % Al + 48 % Ni in the oxidation of CO and C_mH_n decreased after 10–20 hours of work due to poisoning; it appeared unstable towards SO₂. Over the new catalyst and the catalyst promoted with Pt, the presence of SO₂ causes a significant shift (by 200 °C) of the curves of oxidation to the higher temperature region.

Complete oxidation of CO and 95 % C_mH_n in the presence of SO_2 was observed at temperatures of 450 °C, over the new catalyst, and 400–420 °C, over the Pt-catalyst at $W = 50 \times 103 \text{ m}^3 / \text{m}^3 \cdot \text{h}$. The new catalyst worked for 600 hours.

The comparison of the results showed that in the conditions of thermal autoignition the new catalyst has substantial advantages with respect to the contacting

temperature since it allows for temperatures of the process lower by ≈ 80 °C. It has 1.6–1.7 times longer duration of operation than the catalyst with the composition of 52 % Al + 48 % Ni, and its catalytic properties are equal to those of the catalysts containing noble metals. Based on the results we recommend the catalyst for industrial use.

For neutralization of exhaust gases from kilns, we recommend the two-step purification scheme, according to which the first step is the purification of the gas by C-type electrostatic precipitator from resinous substances and the second step is the elimination of carbon monoxide in the catalytic reactor (Fig. 2). We recommend the one-step neutralization of the waste gas in the catalytic reactor for graphitization furnaces (Fig. 3). Each kiln and graphitization furnace needs its own reactor.

By means of a ventilation unit, the exhaust gases produced by kilns and graphitization furnaces are sent to the preheater, where they are heated by the heat of flue gases produced by natural gas burning to the temperature of 200 °C to start the catalytic reaction, and then pass to the catalytic reactor for neutralization. We recommend using the new intermetallic catalyst with the composition of 30 % Ni + 10 % Co + 11 % Mn + 2 % Cu + 47 % Al [9].

The most compact and economical device is a cyclone-type reactor with a radial gas inlet. The catalyst basket and the adjacent tube-type heat exchanger are inside [10]. Between the reactor shell and the outer shell of the catalyst basket, there is an annular channel through which the gas flows. In such a reactor, two processes take place simultaneously: the catalytic oxidation of CO and C_mH_n in the catalyst bed and the heat recovery. First, the exhaust gas flows through the shell side of the recuperator, where it absorbs heat

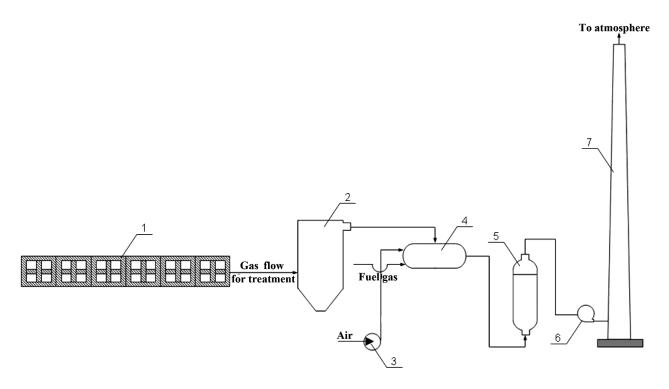


Fig. 2. Installation diagram of catalytic neutralization of kiln waste gases:

1-kiln; 2-electrostatic precipitator of resinous substances; <math>3-ventilator; 4-furnace-preheater; 5-reactor; 6-smoke exhauster; 7-flue gas stack

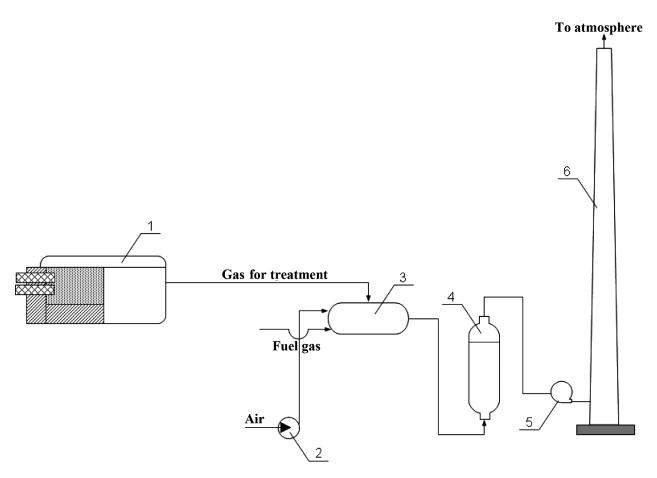


Fig. 3. Installation diagram of catalytic neutralization of graphitization furnace waste gases: 1- graphitization furnace; 2- ventilator; 3- furnace-preheater; 4- reactor; 5- smoke exhauster; 6- flue gas stack

from the partially purified exhaust gas, next, to the header and the mixing chamber, where it is mixed with the hot flue gases fed from the combustion chamber. The exhaust gases heated to the temperature required for the catalytic oxidation are headed to the filling layer of the catalyst basket for oxidizing CO and C_mH_n to carbon dioxide and water. The purified hot exhaust gases flow to the tube side of the recuperator and the smoke exhauster removes it from the apparatus to the flue gas stack.

The total gas flow rate through the reactor was 40000 m³/hr, the required amount of catalyst was 1.25 m³ in the catalyst bed of 0.44 m height, the catalyst weight was 540 kg. According to GOST 9617-67, the diameter of the reactor is 1.9 m and the height is 3.8 m.

The capital costs of construction of such reactors are usually 15-20 % lower. The combined reactor requires the area of $60-70 \text{ m}^2$, while industrial reactor of the same productivity ($40\ 000\ \text{m}^3/\text{h}$) requires minimum $120\ \text{m}^2$.

The utilization of the new catalyst ensures complete neutralization (99.9 %) of CO and C_mH_n in the kiln exhaust gas at the temperature of 300° C with the volumetric flow rate of 32 000 h⁻¹, which ensures low energy consumption of the process. A higher temperature is required (450 °C) to achieve the same effect in the neutralization of the exhaust gas of graphitization furnaces.

In the process of purification of exhaust gas of the kiln, the new catalyst worked for 720 hours and in the graphitization furnace, it worked for 600 hours. When the activity of the catalyst reduced to 95 % because of the carburization with resinous substances, it subjected to regeneration.

The catalyst with the new composition can be used at different plants for neutralization of CO and C_mH_n .

To assess whether the air in the housing area affected by the emissions of the PAT "Ukrgrafit" meets

the sanitary standards the ground level concentrations of pollutants were measured before and after installation of the catalytic gas purification plant for kilns and graphitization furnaces. The calculation was performed using the PLEINAIR-1.25 software developed by the SP "Interex" (Kyiv, 1994). The software works in accordance with the document OHД-86 "Method of Calculation of the Harmful Substances Concentration in the Air Polluted by Industrial Emissions" and was approved by the Ministry of Ecology and Natural Resources of Ukraine.

The modelling of the dispersion of pollutants in the air of the residential area, taking into account the background level, showed the elevated surface concentrations of carbon monoxide, naphthalene and phenol of 1.05 MAC, 1.1 MAC and 1.14 MAC, respectively. The results proved the need for conservation measures to minimize emissions of these substances.

Currently, according to Order no. 309, the emissions of all harmful substances produced by the kilns and graphitization furnaces of PAT "Ukrgrafit" do not exceed norms, except for carbon monoxide. The concentration of carbon monoxide in the emissions 2–7 times exceeds the norm (the maximum concentration is 1750 mg/m³ while the norm is 250 mg/m³).

After neutralization of the waste gases of the kilns and graphitization furnaces over the developed catalyst, the surface concentrations of the pollutants emitted from these sources in residential areas will lower to the background level (Table). The regulations stated in the Order № 309 dated June 27, 2006, will be observed.

Conclusions. The method of deactivation of waste gases containing carbon monoxide and hydrocarbons that are emitted by graphitization furnaces, which is based on intermetallic catalysts, have been developed and recommended for implementation at the enterprise PAT "Ukrgrafit". The results of comparative tests showed that the developed catalyst cycle life is

Table

The values of surface concentrations of pollutants in residential areas before and after the PAT "Ukrgrafit"

exhaust gas purification by the developed catalyst

Substance	Short-Term MAC, mg/m ³	Background Concentration		Maximum Concentration Including Background Concentration Before Purification		Maximum Concentration Including Background Concentration After Purification	
		mg/m ³	MAC fraction	mg/m ³	MAC fraction	mg/m ³	MAC fraction
Carbon monoxide	5.0	4.69	0.938	0.571 (5.26)	0.114 (1.052)	0.0003 (4.69)	$7 \cdot 10^{-5} (0.938)$
Benzol	1.5	0.6	0.4	0.018 (0.618)	0.012 (0.412)	0.00003 (0.60)	$2 \cdot 10^{-5} (0.4)$
Styren	0.04	0.016	0.4	0.011 (0.027)	0.280 (0.680)	0.00005 (0.016)	0.001 (0.401)
Benzapyrene	_	_	_	0.000010 (-)	0.990 (-)	Calculation makes no sense	
Naphthalene	0.003	0.0012	0.4	0.002 (0.003)	0.695 (1.095)	0.00004 (0.0016)	0.012 (0.412)
Acenaphthene	0.07	_	_	0.063 (-)	0.902 (-)	Calculation makes no sense	
Phenol	0.01	0.004	0.4	0.007 (0.011)	0.740 (1.140)	0.00007 (0.0041)	0.007 (0.407)

1.6—1.7 times longer than that of the known nickel catalyst. After neutralization of exhaust gases emitted by graphitization furnaces by means of the developed catalyst, the surface concentrations of the pollutants in residential areas will be at the background levels. The emissions of all contaminants will meet the standards stated in the Order no. 309 dated June 27, 2006.

References/Список літератури

1. Kozhemyakin, G. B., Belokon, K. V. and Rumyantsev, V. R., 2011. Strategy for air quality in the European Union and Ukraine. In: ZDIA, 7th Ukrainian Scientific-Practical Conf. "Okhorona navkolyshnoho seredovyshcha promyslovykh rehioniv yak umova staloho rozvytku Ukrainy". December 15, 2011, Zaporizhzhia: ZDIA, pp. 10–13.

Кожемякин Г.Б. Стратегия обеспечения качества воздуха в Европейском союзе и Украине: матеріали VII Всеукр. наук.-практ. конф. (15 груд. 2011 р., Запоріжжя) / Г.Б. Кожемякин, К. В. Белоконь, В. Р. Румянцев // Охорона навколишнього середовища промислових регіонів як умова сталого розвитку України: зб. статей — Запоріжжя: ЗДІА, 2011. — С. 10—13.

2. Kozhemyakin, G. B. and Belokon, K. V., 2012. The reduction of the admission carbonaceous components of industrial emissions into the atmosphere through the use of intermetallic catalysts. In: UkrINDIEP, 8th Int. Scientific-Practical Conf. "Ekolohichna bezpeka: problemy i shliakhy vyrishennia". 10–14 September 2012, Alushta: UkrINDIEP, Vol. 2, pp. 177–180.

Кожемякин Г.Б. Снижение поступления углеродсодержащих компонентов промышленных выбросов в атмосферу путем использования интерметаллидных катализаторов: матеріали VIII міжнар. наук.-практ. конф. 10—14 верес. 2012 р., Алушта / Г.Б. Кожемякин, К.В. Белоконь // Екологічна безпека: проблеми і шляхи вирішення: зб. наук. статей. — Харків, УкрНДІЕП, 2012. — Т. 2. — С. 177—180.

3. Vinogradov, S.S. and Vasilyeva, I.A., 2007. Methods and equipment for cleaning and disinfection of air emissions. *Ekologiya Proizvodstva*, No. 1, pp. 39–43.

Виноградов С. С. Методы и оборудование для очистки и обезвреживания выбросов в атмосферу / С. С. Виноградов, И. А. Васильева // Экология производства. -2007. - №1. - C. 39-43.

4. Ladygichev, M.G. and Berner, G.Ya., 2004. *Zarubezhnoe i otechestvennoe oborudovanie dlya ochist-ki gazov* [Foreign and domestic gas purification equipment], Moscow: Teplotehnik.

Ладыгичев М. Г. Зарубежное и отечественное оборудование для очистки газов / Ладыгичев М. Г., Бернер Г. Я. — М.: Теплотехник, 2004. — 696 с.

5. Romanov, I. V., Farbun, I. A., Khaynakov, S. A., Kirillov, S. A. and Zazhigalov, V. A., 2008. Investigation of the catalytic properties of materials based on transition metal oxides and cerium. *Dopovidi Natsionalnoi Akademii Nauk Ukrainy*, No. 10, pp. 154–159.

Исследование каталитических свойств материалов на основе оксидов переходных металлов и церия / И.В. Романова, И.А. Фарбун, С.А. Хайнаков [и др.] // Доповіді Національної академії наук України. — 2008. - N 10. - C. 154-159.

6. Borsch, V. N., Pugacheva, Ye. V., Zhuk, S. I, Andreyev, D. Ye, Sanin, V. N. and Yukhvid, V. I., 2008. Multi-metal catalysts of deep oxidation of CO and hydrocarbons. *Doklady Akademii Nauk (RF)*, Vol. 419, No. 6, pp. 775–777.

Многокомпонентные металлические катализаторы глубокого окисления СО и углеводородов / В. Н. Борщ, Е. В. Пугачева, С. Я. Жук [и др.] // Доклады Академии Наук РФ. — 2008. — № 419 (6). — С. 775—777.

- **7.** Sereda, B., Belokon, Y., Zherebtsov, A. and Sereda, D., 2012. The researching and modeling of physical-chemical properties of Ni-base alloys in SHS conditions. *Materials Science and Technology*, Pittsburgh, Vol. 1, pp. 494–498.
- **8.** Sereda, B., Zherebtsov, A., Kruglyak, I., Belokon, Y., Savela, K. and Sereda, D., 2010. The retrieving of heat-resistant alloys on intermetallic base for details of gas turbine engine hot track in SHS conditions. *Materials Science and Technology*, Houston, Vol. 3, pp. 2097–2102.
- **9.** Sereda, B. P., Savela, K. V., Kozhemyakin, G. B., Belokon, Yu. A. and Ryzhkov, V. G., Zaporizhzhian State Engineering Academy, 2009. *Katalizator dlia ochyshchennia oksydu vuhletsiu i vuhlevodniv* [The catalyst for the purification of carbon monoxide and hydrocarbons], Patent of Ukraine, no. 45154.

Пат. 45154 України, МПК В01Ј25/00. Каталізатор для очищення оксиду вуглецю і вуглеводнів / Б. П. Середа, К. В. Савела, Г. Б. Кожемякін [та ін.]; заявник і патентоволодарь — Запоріж. держ. інж. академія. № и 2009 05588; заявл. 01.06.2009; опубл. 26.10.2009, Бюл. № 20.

10. Batura, P. I., 1991. Catalytic reactors for afterburners flue gas. *Koks i Khimiya*, No. 5, pp. 32–34.

Батура П. И. Каталитические реакторы для дожигания отходящих газов / П. И. Батура // Кокс и химия. — 1991. - №5. - C. 32-34.

Мета. Удосконалення процесів знешкодження газових викидів підприємств виробництва електродів для забезпечення якості атмосферного повітря та дотримання екологічних нормативів, встановлених в Україні.

Методика. Випробування каталізаторів на ефективність проводили у струмі відхідних газів печей випалу та графітації на лабораторній установці при варіюванні температури від 100 до 500 °C і об'ємної швидкості газового потоку W від $30 \cdot 10^3$ до $120 \cdot 10^3$ м³/м³ · год. Для оцінки дотримання санітарно-гігієнічних норм у житловій забудові в зоні дії викидів до і після установки каталітичного очищення газів, що відходять від печей випалу та графітації, виконано розрахунок рівня приземних концентрацій забруднюючих речовин в атмосферному повітрі з використанням про-

грамного комплексу "ПЛЕНЕР-1.25", заснованого на "Методики розрахунку концентрацій в атмосферному повітрі шкідливих речовин, що містяться у викидах промислових підприємств" (ОНД-86) і погодженого з Мінприроди України.

Результати. Моделювання розсіювання забруднюючих речовин у повітрі показало перевищення приземних концентрацій у житловій зоні з урахуванням фону для оксиду вуглецю, нафталіну і фенолу, які становлять, відповідно, 1,05 ГДК, 1,1 ГДК і 1,14 ГДК, що зумовило необхідність проведення природоохоронних заходів з мінімізації викидів цих речовин. У роботі проведено порівняльні випробування з використанням розробленого та існуючого каталізаторів для знешкодження відхідних газів від печей випалу та графітації. Результати порівняльних випробувань показали, що розроблений склад каталізатора має термін служби в 1,6–1,7 рази більший, ніж у відомого нікелевого каталізатора. Застосування розробленого складу каталізатора дозволило підвищити ступінь очищення газів до 99,9 %. Після знешкодження технологічних газів від печей випалу та графітації на розробленому каталізаторі, приземні концентрації всіх забруднюючих речовин, що відходять від цих джерел, у житловій забудові будуть на рівні фонових значень.

Наукова новизна. Теоретично обґрунтовано та експериментально підтверджено підвищення екологічної безпеки викидів шляхом каталітичного знешкодження оксиду вуглецю та вуглеводнів на інтерметалідних каталізаторах.

Практична значимість. Розроблено та рекомендовано для впровадження на підприємстві ПАТ "Укрграфіт" спосіб знешкодження газових викидів, що містять оксид вуглецю й вуглеводні, від печей випалу та графітації на інтерметалідних каталізаторах.

Ключові слова: газові викиди, оксид вуглецю, вуглеводні, знешкодження, інтерметалідний каталізатор

Цель. Совершенствование процессов обезвреживания газовых выбросов предприятий производства электродов для обеспечения качества атмосферного воздуха и соблюдения экологических нормативов, установленных в Украине.

Методика. Испытания катализаторов на эффективность проводили в токе отходящих газов печей обжига и графитации на лабораторной установке при варьировании температуры от 100 до 500 °C и объемной скорости газового потока W от $30 \cdot 10^3$ до $120 \cdot 10^3$ м 3 /м 3 ·ч. Для оценки соблю-

дения санитарно-гигиенических норм в жилой застройке в зоне действия выбросов до и после установки каталитической очистки отходящих газов от печей обжига и графитации выполнен расчет уровня приземных концентраций загрязняющих веществ в атмосферном воздухе с использованием программного комплекса "ПЛЕ-НЭР-1.25", основанного на "Методике расчета концентраций в атмосферном воздухе вредных веществ, содержащихся в выбросах промышленных предприятий" (ОНД-86) и согласованного с Минприроды Украины.

Результаты. Моделирование рассеивания загрязняющих веществ в воздухе показало превышение приземных концентраций в жилой зоне с учетом фона для оксида углерода, нафталина и фенола, которые составляют соответственно 1,05 ПДК, 1,1 ПДК и 1,14 ПДК, что обусловило необходимость проведения природоохранных мероприятий по минимизации выбросов этих веществ. В работе проведены сравнительные испытания с использованием разработанного и существующего катализаторов для обезвреживания отходящих газов от печей обжига и графитации. Результаты сравнительных испытаний показали, что разработанный состав катализатора имеет срок службы в 1,6-1,7 раза больший, чем у известного никелевого катализатора. Применение разработанного состава катализатора позволило повысить степень очистки газов до 99,9 %. После обезвреживания технологических газов от печей обжига и графитации на разработанном катализаторе, приземные концентрации всех загрязняющих веществ, отходящих от этих источников, в жилой застройке будут на уровне фоновых значений.

Научна новизна. Теоретически обосновано и экспериментально подтверждено повышение экологической безопасности выбросов путем каталитического обезвреживания оксида углерода и углеводородов на интерметаллидных катализаторах.

Практическая значимость. Разработан и рекомендован для внедрения на предприятие ПАО "Укрграфит" способ обезвреживания газовых выбросов, содержащих оксид углерода и углеводороды, от печей обжига и графитации на интерметаллидных катализаторах.

Ключевые слова: газовые выбросы, оксид углерода, углеводороды, обезвреживание, интерметаллидный катализатор

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