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ЕКОЛОГІЯ ТА РЕСУРСОЗБЕРЕЖЕННЯ

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BLOCK CATALYTIC SYSTEM FOR NEUTRALIZATION OF CARBON MONOXIDE BASED ON AERATED CONCRETE

The paper presents the results of a study of catalysts for the conversion of carbon monoxide based on aerated concrete, modified with magnetite and chromium ferrite separately and in aggregate. It was found that at a consumption of 100 g of catalyst powder per 1 dm3 of a typical mixture for producing aerated concrete and obtaining blocks of modified aerated concrete according to the traditional technology, their efficiency is 70-85% at 400 °C and decreases to 9-13% at 200 °C. In terms of strength and physicochemical properties, aerated concrete samples differ little from standard ones, and in some cases even exceed them. The proposed method for fixing catalyst particles in blocks of aerated concrete makes it possible to build fundamentally new schemes for neutralizing carbon monoxide when placing modified blocks directly at the loading of electrode raw materials in furnaces. This greatly simplifies the conversion process and its control system.

***Keywords**: carbon monoxide, flue gases, purification, catalyst, oxidation, conversion, multi-chamber furnaces, aerated concrete*

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Problem statement. Environmental pollution is increasing in scale every year and requires increased attention to the problems that arise in this case. The atmosphere, as the most mobile component of the environment and the most voluminous resource for the existence of living organisms, especially suffers from anthropogenic pollution. And if even schoolchildren know about atmospheric pollution with carbon dioxide and the greenhouse effect, then such a pollutant as carbon monoxide (CO) has not yet received due attention from both scientists and industrial workers. At the same time, it is well known that carbon monoxide is a dangerous gas for living organisms and can cause suffocation, hypoxia, and even death [1]. Most of all poisoning in the world is associated with carbon monoxide intoxication. On the other hand, CO emissions are associated with the operation of the metallurgical and petrochemical industries, with the operation of furnaces for roasting and graphitization furnaces of electrode products, and with the production of many chemical compounds, plastics, and automobile fuel [2, 3, 4]. In the emissions of many enterprises, the CO content can reach 20% and can increase to 40 - 80% for some processes [5].

Analysis of previous studies. Among all the currently known methods for the conversion of carbon monoxide, the most acceptable and widespread is the catalytic method. This method is most widely used in the neutralization of emissions from road transport [6]. In industry, large-scale installations for the conversion of carbon monoxide are still practically absent. And this is largely due to the cost of materials used in catalysts - palladium, platinum, rhodium [7, 8]. In addition, significant problems arise during the manufacture of the catalyst itself. On the one hand, the intensity of the catalytic process depends on the contact area between the catalyst and the gas phase. On the other hand, with a significant dispersion of the solid phase, the devices for keeping the catalyst particles in the working volume of the equipment become much more complicated. Therefore, catalysts are quite often applied to a granular mineral support [9, 10]. However, even in this case, the handling of the catalyst – the formation of the working space, regeneration, disposal, etc., does not become more convenient and cheaper. The problem of fixing a highly dispersed catalyst in a certain volume remains completely unsolved.

The current state of research in the field of production of building materials shows that the addition of a certain part of mineral additives, including zeolites with sorbed heavy metals, to aerated concrete does not significantly affects its properties, and in some cases even allows to save cement with a simultaneous increase in the strength of products obtained from it [11, 12]. In this case, such a step also makes it possible to resolve the issue of utilization of spent metal oxide catalysts for CO oxidation of flue gases from electrode production [9]. Therefore, let's propose to use aerated concrete, which has a developed porous structure, as a fixer for catalyst particles. This makes it possible not only to ensure maximum contact between the reacting media, but also to significantly simplify the formation of the working space in real installations for neutralizing carbon monoxide.

The aim of this research is to study the main mechanical and physicochemical properties of ferrite catalysts based on aerated concrete and their efficiency in the processes of catalytic oxidation of carbon monoxide with atmospheric oxygen. This will make it possible to calculate a catalytic CO oxidation reactor of a new design.

Materials and methods.

Obtaining a catalyst. Catalyst samples for the oxidation of carbon monoxide from gas emissions were obtained by forming a mixture for industrial production of aerated concrete with the addition of catalyst powder. The main characteristics of the mixture are given in Table 1.

Table 1 – Chemical composition and characteristics of a typical mixture for the production of aerated concrete

№	Parameter	Units	Value
1	Density	kg dm^{-3}	1.69
2	Dry matter content	kg m^{-3}	460.0
3	Binder content:	%	31.4
4	- Incl. cement АШ 500	%	76.6
5	- Incl. quicklime CaO	%	23.4
6	Gypsum content $CaSO_4 \cdot 2H_2O$	%	3.78
7	Content of aluminum paste (aluminum content in the paste is 70%)	%	0.14
8	Sand content	%	64.68

In the process of preparing a mixture for the industrial production of aerated concrete, sand with a grain size of 8 µm was used, to which water, cement, lime, gypsum were added in portions, mixed, aluminum paste was added, and the final mixing was carried out.

To obtain catalyst samples up to 1 dm3 of the finished mixture for aerated concrete production, 100 g of catalyst powder was added, the components were thoroughly mixed, and the mixture was poured into molds with a size of 100 × 100 × 100 mm. Then the molds were kept at a temperature of 50 °C for 5 hours. Then the molds were transferred to an autoclave and kept at a temperature of 180 °C for 10 hours. As a result of the described process, catalyst samples for the oxidation of carbon monoxide were obtained.

Three samples were used as catalyst powders:

1. Chromium-ferrite catalyst $Fe(Fe_xCr_{1-x})_2O_4$ ($Cr^{6+}:Fe^{2+}=1:15$). Solutions of 0.232M $K_2Cr_2O_7$ and 2.756M $FeSO_4$ were poured into one container and precipitated with 25% NH_4OH solution following the reactions:

$$Cr_2O_7^{2-} + 2OH^- \rightarrow 2CrO_4^{2-} + H_2O, \tag{1}$$

$$2K_2CrO_4 + 9Fe(OH)_2 \rightarrow 3Fe_3O_4\downarrow + 2Cr(OH)_3\downarrow + 4KOH + 4H_2O. \tag{2}$$

The resulting suspension was settled for 1 hours, the solid particles were separated from the liquid phase by decantation, washed until neutral, dried in air to constant weight, and ground to form particles with a size of 0.25 mm.

2. Magnetite Fe_3O_4. 1 dm^3 of 1M $FeSO_4$and 2M $FeCl_3$ solutions were prepared each, they were poured off and precipitated with 25% NH_4OH solution. As a result, a suspension of magnetite particles was obtained according to the reaction:

$$FeSO_4 + 2FeCl_3 + 8NH_4OH \rightarrow Fe_3O_4\downarrow + 4H_2O + (NH_4)_2SO_4 + 6NH_4Cl. \tag{3}$$

The suspension was settled in the mother liquor for 1 hours, the solid magnetite particles were separated from the liquid phase by decantation, washed until neutral, dried in air to constant weight, and ground to form particles with a size of 0.25 mm.

3. Mixture 50 % $Fe(Fe_xCr_{1-x})_2O_4$ (Cr:Fe=1:15), 50 % Fe_3O_4. Powders were obtained by the reactions described above and mixed in equal amounts.

To determine the mechanical properties of the obtained catalyst samples based on aerated concrete, an IP-100 hydraulic press was used.

Isotherms of low-temperature nitrogen adsorption for all synthesized samples obtained by the volumetric method using Sorptomatic 1990 Sorbtometers at the boiling point of liquid nitrogen (77 K). All samples were pre-degassed at 300 °C in a helium flow. The volume of adsorbed nitrogen was determined at the time of equilibrium in the nitrogen flow.

The value of the specific surface area S_0 was calculated according to the Brunauer-Emmett-Teller (BET) theory [13] using the formula [14]:

$$S_0=a_m\cdot N_A\cdot\omega, \tag{1}$$

where a_m – limiting capacity of an adsorbate monolayer, N_A – Avogadro's constant, and ω – surface area occupied by one adsorbed molecule (0.162 nm^2 for nitrogen [15]). The value of the limiting capacity of the monolayer was calculated from the linear BET equation [16]:

$$\frac{\left(\frac{P}{P_0}\right)}{a\cdot\left(1-\frac{P}{P_0}\right)}=\frac{1}{a_m}+\frac{C-1}{a_m\cdot C}\cdot\frac{P}{P_0}, \tag{2}$$

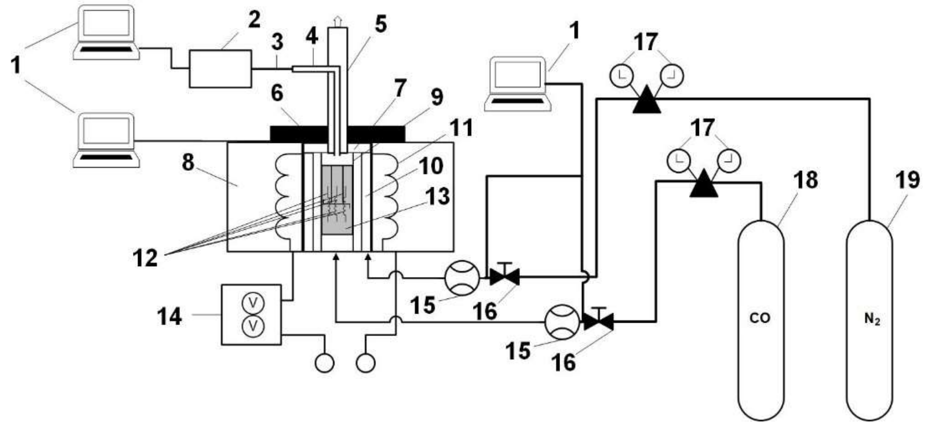
where C – constant. The linearization of the experimental data of the isotherm in the range of relative pressure $P/P_0 = 0.2–0.4$ in the corresponding coordinate system according to equation (2) makes it possible to determine the numerical values of am and C. Such an algorithm for calculating the specific surface area is implemented in the software of the Sorptomatic 1990 device, which was used for processing experimental data in this work.

The average pore size was determined based on the Barrett-Joyner-Halenda (BJH) method [17] according to the Thompson-Kelvin equation [18]:

$$ln\frac{P}{P_0}=-\frac{2\cdot V_m\cdot\sigma}{r\cdot R\cdot T}, \tag{3}$$

The study of the efficiency of the catalytic conversion of carbon monoxide by catalysts based on aerated concrete was carried out using a laboratory setup shown in Fig. 1 [10]. The main element of the installation is a heat chamber 8. It is a stainless steel tube 9, consisting of a metal tube made of stainless steel 9, which is inserted into a ceramic tube 10. The latter contains a heating element 11 to maintain a given temperature in the heat chamber. The operation of the heating element is controlled by thermocouples 12 and a thermostat 14. A sample of aerated concrete with a catalyst in a cylindrical container 13 was placed in a metal tube 9. The dimensions of the sample were: height 160 mm, diameter 56 mm. The catalyst was placed in tube 9 without gaps and slits.

The study of the efficiency of oxidation of carbon monoxide was carried out in a flow-through unit at atmospheric pressure. The composition of the gas mixture was constantly monitored at the entrance and exit from the heat chamber. The content of the reaction products was analyzed using a gas analyzer 2 with an accuracy of ± 20 ppm or ± 5% of the measurement values. The oxidation process of carbon monoxide was carried out at different volumetric gas flow rates in the range of 1-5 dm^{3}min^{-1}. The mole fraction of carbon monoxide in the initial gas mixture at the inlet to the reactor is 1.1. The catalyst was tested under the same conditions in the temperature range 200–400 °C. Nitrogen was used as an inert gas, if necessary. Before carrying out the catalytic experiment, a sample of the catalyst was heat treated at 450 °C for 2 h.



1 - digital measurement interface; 2 - gas analyzer VARIO PLUS industrial analyzer MRU air fair; 3 - gas analyzer sampler; 4 - tube for sampling the gas mixture from the chamber to the gas analyzer; 5 - outlet of the gas mixture; 6 - metal gasket; 7 - asbestos gasket; 8 - heat chamber; 9 - stainless steel tube; 10 - ceramic tube; 11 - heating element; 12 - thermocouples; 13 - container for catalyst made of stainless steel mesh; 14 - temperature regulator; 15 - gas flow meters; 16 - control valves; 17 - reducers; 18 - carbon monoxide cylinder; 19 - nitrogen cylinder

Fig. 1 – Diagram of a laboratory setup for studying the processes of catalytic oxidation of carbon monoxide

The catalytic activity of aerated concrete samples in the oxidation of carbon monoxide was characterized by the conversion of CO to CO_2.

Carbon monoxide conversion was calculated using the formula:

$$X_{CO} = \frac{C_{CO}^{in} - C_{CO}^{out}}{C_{CO}^{in}} \cdot 100\%, \tag{4}$$

where $C_{in}CO$ – CO molar fraction at the inlet to the reactor, $C_{out}CO$ – molar fraction of CO at the outlet of the reactor.

Presentation of the main material. Investigation of the mechanical characteristics of aerated concrete mixture with and without catalyst showed satisfactory results (Table 2). At 100 g of powder per 1 dm^3 of aerated concrete mixture, the decrease in the strength of the cubes obtained from them was 2.64-11.21%. However, a 5.28% increase in strength was also observed with the addition of a chromium ferrite catalyst. Thus, the addition of catalyst powder to the aerated concrete mixture does not critically affect the properties of products, that is, products during long-term operation in production will not be subject to destruction.

Table 2 – Dependence of the compressive strength of the obtained aerated concrete cubes on the chemical composition of the catalyst powder

No. of catalyst powder	Chemical composition of catalyst powder	Strength of cubes, MPa
Without catalyst	-	3.03
1	Fe_3O_4	2.95
2	$Fe(Fe_xCr_{1-x})_2O_4$	3.19
3	50 % $Fe(Fe_xCr_{1-x})_2O_4$, 50 % Fe_3O_4	2.69

A study of the porous properties of products made of aerated concrete modified with a catalyst powder showed (Figs. 2, 3) that the adsorption isotherms obtained for samples of aerated concrete modified with magnetite and chromium ferrite (calcined and uncalcined) belong to type II isotherms, concave relative to the P/P_0 axis and rather steep bend [17]. The isotherms show long rectilinear sections, the start points of which correspond to the relative pressure at which the adsorption of the monolayer is completed. Type II isotherms are inherent in macroporous adsorbents, which is fully confirmed by the results of calculating the parameters of the porous structure of modified aerated concrete given in Table 2.

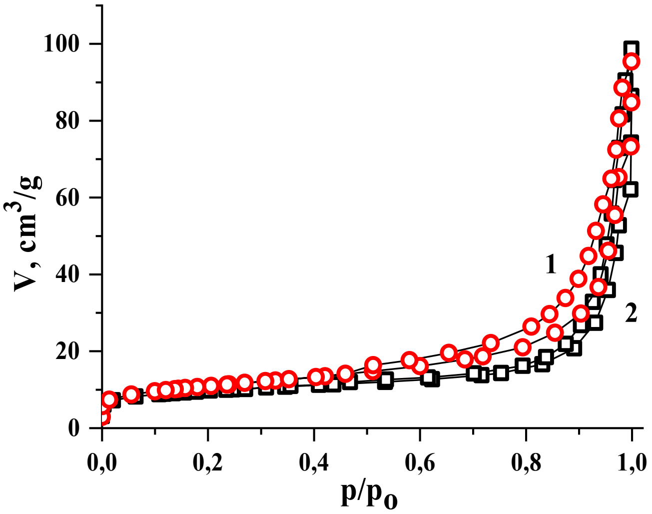


Fig. 2 – Isotherms of nitrogen adsorption and desorption on samples of unfried (1) and fried (2) aerated concrete modified with chromium ferrite

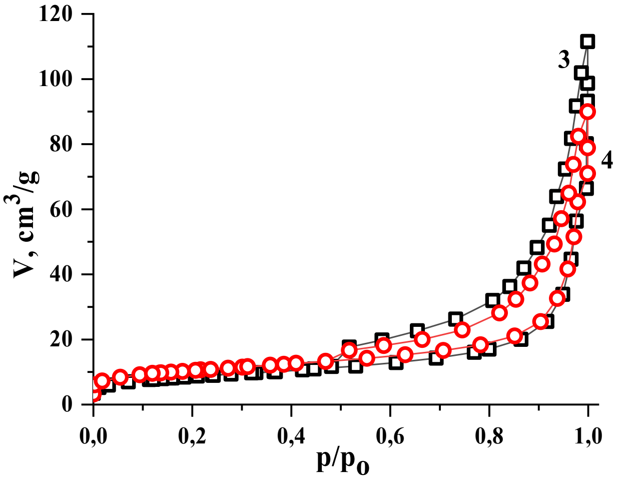


Fig. 3 – Isotherms of nitrogen adsorption and desorption on samples of unfried (3) and fried (4) aerated concrete modified with magnetite

As can be seen from Table 3, heat-treated and thermally non-refined aerated concrete modified with magnetite, and thermo-non-refined aerated concrete modified with chromium ferrite have approximately the same insignificant content of micropores, which is 5.22-5.85%. At the same time, heat-treated aerated concrete modified with chromium ferrite has a 2 times higher content of micropores. This fact can be explained by the fact that the synthesized magnetite by the water condensation method has a more ordered crystal structure than chromium ferrite. Even in the case of the optimal ratio Cr^{6+}:Fe^{2+}=1: 15 [20], the formation of an amorphous phase of iron hydroxide around crystalline particles of chromium ferrite with a high content of hydrated water is possible, it did not evaporate even as a result of heat treatment in an autoclave at 190 °C for 10 hours. A decrease in the pore size in this case by almost 2.5 times indicates the formation of a more ordered crystal structure of chromium ferrite under the action of an elevated temperature, which is also confirmed by a slight decrease in the specific surface area of the aerated concrete sample modified with chromium ferrite.

Table 3 - Characteristics of the porous structure of modified aerated concrete

№	Sample	Specific surface by BET S, m^2g^{-1}	Total pore volume V_Σ, cm^3g^{-1}	Micropore volume V_μ, cm^3g^{-1}	Micropore content $^*V_{\mu\%}$, %	Pore size distribution, nm	
						BJH dV (r), nm	
						r_1	r_2
1	Thermal non-refurbishment of aerated concrete, modification with chromium ferrite	39.312	0.1020	0.0057	5.58	3.92	-
2	Heat curing of aerated concrete, modification with chromium ferrite	34.795	0.082	0.0095	11.58	1.65	-
3	Thermal non-refining of aerated concrete, modification with magnetite	31.258	0.0881	0.0046	5.22	4.05	-
4	Thermal curing of aerated concrete, modification with magnetite	37.892	0.1282	0.0075	5.85	4.00	-

$^*V_{\mu\%} = V_\mu/V_\Sigma \cdot 100\%$.

A study of the efficiency of CO conversion on samples of aerated concrete modified with chromium ferrite and magnetite showed that they provide approximately the same conversion rate (Fig. 4). Magnetite stands out somewhat in terms of efficiency; in our opinion, it may be associated with the greater formation of its particles and, accordingly, an increase in the contact area between the reacting media. And if chromium ferrite provides the maximum degree of CO conversion in the temperature range up to 320 °C, then magnetite is most effective at high temperatures. For a mixture of magnetite and chromium ferrite, then when using it, the degree of conversion of carbon monoxide approaches the efficiency of magnetite at temperatures above 375 °C.

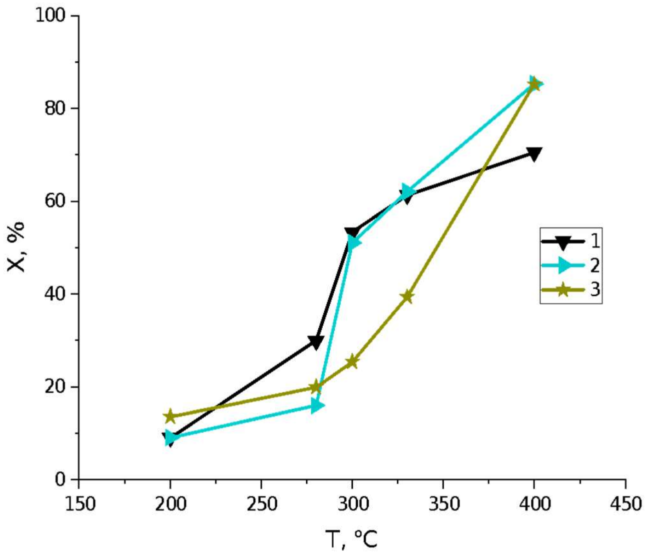


Fig. 4 – Efficiency of CO conversion on various samples of modified aerated concrete, depending on temperature: 1 – chromium ferrite; 2 - magnetite; 3 – mixture of chromium ferrite and magnetite (50:50)

The use of aerated concrete for fixing the catalysts for the CO conversion can significantly simplify the very design of the system for neutralizing carbon monoxide. Today, most often, for example, in furnaces for roasting electrode products, containers with a catalyst are placed in the firing channels of chambers heated by flue gases [9].

In this case, in the manufacture of blocks with a catalyst with dimensions of 600 × 200 × 100 mm from aerated concrete, they can be laid out directly on a thermo-anthracite pouring "green" blanks in furnaces for firing electrode products (Fig. 5, 6, 7).

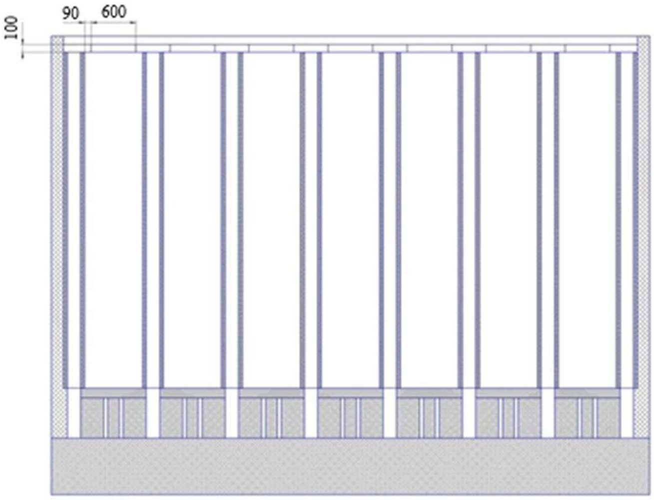


Fig. 5 – Placing aerated concrete blocks with a catalyst directly on the surface of the pouring of electrode blanks in firing furnaces (front view)

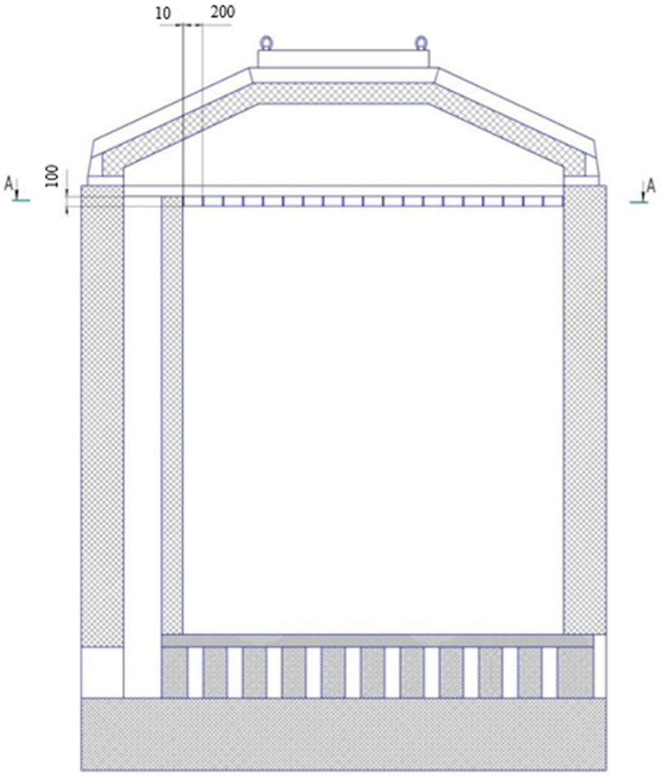


Fig. 6 – Placement of aerated concrete blocks with a catalyst directly on the surface of pouring electrode blanks in firing furnaces (side view)

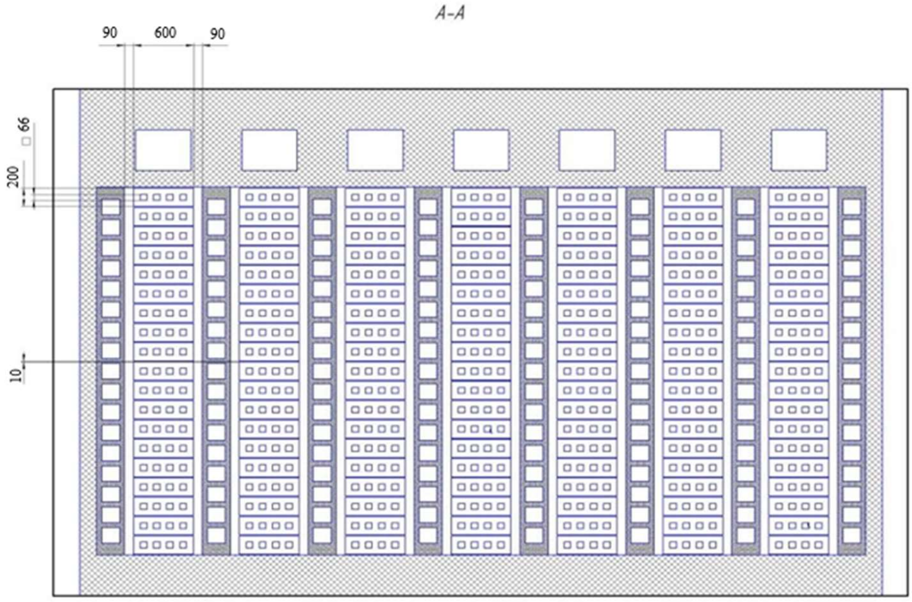


Fig. 7 – Placing aerated concrete blocks with a catalyst directly on the surface of the pouring of electrode blanks in kilns (top view)

As can be seen from Fig. 5, 6, 7, the concrete blocks adjoin each other at a distance of 10 mm, while the presence of a sufficient number of holes in the blocks of a square size provides the movement of the gas mixture inside the chamber necessary for the passage of the firing process. Blocks can be made with 4 holes 66 × 66 mm in size (Fig. 8), and with 14 holes 40 × 40 mm (Fig. 9). In the first case, a greater strength of aerated concrete blocks is ensured during operation, in the second, an increase in the specific surface area of the catalyst contact with flue gases containing CO.

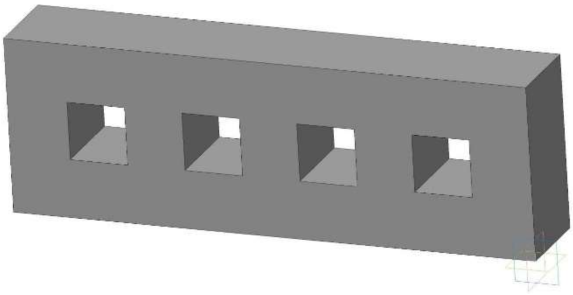


Fig. 8 – Aerated concrete blocks with holes 66 × 66 mm

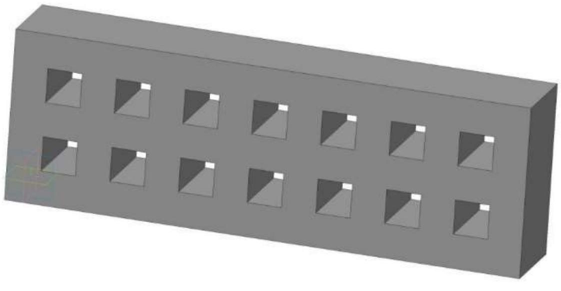


Fig. 9 – Aerated concrete blocks with holes of 40 × 40 mm

CONCLUSIONS

A catalyst modified with chromium ferrite and magnetite based on aerated concrete are developed. The main strength and physicochemical characteristics of the catalyst are established, and its efficiency in the CO conversion processes is determined. It is determined that at a consumption of 100 g of catalyst powder per 1 dm^3 of a typical mixture for producing aerated concrete, their efficiency is 70-85% at 400 °C and decreases to 9-13% at 200 °C.

It is proposed to manufacture the catalyst in the form of blocks with dimensions of 600 × 200 × 100 mm, which makes it possible to radically change the scheme of CO neutralization. Catalysts in this form can be placed in the form of a layer of blocks directly on the loading of electrode products, which greatly simplifies their maintenance system.

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БЛОЧНА КАТАЛІТИЧНА СИСТЕМА ЗНЕШКОДЖЕННЯ МОНООКСИДУ ВУГЛЕЦЮ НА ОСНОВІ ГАЗОБЕТОНУ

Натепер в Україні проблема очищення газів від монооксиду вуглецю стоїть достатньо гостро. Одним з найбільш перспективних методів очищення є каталітичне "допалювання" монооксиду вуглецю киснем повітря, присутнім у самих газах. Запропоновані для цього каталізатори, як правило, містять благородні метали або складні оксиди, при цьому активна речовина каталізатора переважно наноситься на поверхню гранульованого носія з розвиненою поверхнею. Проте каталізатори, що використовуються, мають ряд недоліків: поводження з каталізатором – формування робочого простору, регенерація, утилізація тощо, не стає більш зручним та дешевим. Проблема фіксації високодисперсного каталізатора у визначеному об'ємі залишається не повністю вирішеною. В роботі запропоновано в якості фіксатора частинок каталізатора використовувати газобетон, котрий має розвинену пористу структуру. Це дозволяє не лише забезпечувати максимальний контакт між реагуючими середовищами, а й значно спростити формування робочого простору в реальних установках знешкодження монооксиду вуглецю. Розробка високоефективних каталізаторів з використанням газобетону промислового виробництва в якості фіксатора частинок активної речовини - сполук Fe(II), Fe(III), Cr(III) - і стала метою даної роботи.

В роботі представлені результати дослідження каталізаторів конверсії монооксиду вуглецю на основі газобетону, модифікованих магнетитом та феритом хрому окремо та в сукупності. Встановлено, що при витраті 100 г порошку каталізатора на 1 дм^3 типової суміші для виробництва газобетону та отримання блоків модифікованого газобетону за традиційною технологією їх ефективність складає 70–85 % при 400 °С і знижується до 9–13 % при 200 °С. При цьому за міцністними та фізико-хімічними властивостями зразки газобетону мало відрізняються від стандартних, а в окремих випадках навіть перевищують їх. Запропонований спосіб фіксації частинок каталізатора в блоках газобетону дозволяє будувати принципово нові схеми знешкодження монооксиду вуглецю при розміщені модифікованих блоків безпосередньо на завантаженні електродної сировини в печах. Це суттєво спрощує процес конверсії та систему управління ним, що свідчить про перспективність використовуваного носія.

Ключові слова: *монооксид вуглецю, димові гази, очищення, каталізатор, окиснення, конверсія, багатокамерні печі, газобетон*

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