

## DETERMINATION OF THE CONDITIONS FOR CARBON MATERIALS OXIDATION WITH CARBON MONOXIDE FORMATION AT HIGH TEMPERATURES

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**Abstract.** In this paper, the influence of carbon material type, temperature and oxygen concentration in gas mixture on the processes of carbon monoxide formation in production of the electrodes by graphitization was explored experimentally. Specific quantity of gas formed for a definite time, reduced to mass unit of carbon loading using pitch, packing materials and charge mixture of industrial use, was calculated. It is demonstrated that pitch provides the highest rate of carbon oxidation with the release of CO and substantially exceeds packing materials and charge mixture for this index.

**Keywords:** carbon monoxide, gas release, packing materials, pitch, charge mixture.

### 1. Introduction

Operation of a modern industrial enterprise should be in accordance with the principles of sustainable development, which, in turn, is guided by compliance with requirements of environmental protection, in particular, the control of emission of flue gases. At present, ‘Ukrgrafite’, PrJSC, that ranks high in the domestic industry as a supplier of electrode products not only for Ukrainian industry but for foreign consumers too, pays considerable attention to the issue of reducing the technological impact on the environment [1].

For decades, the Company has been systematically monitoring composition of the flue gases, the main harmful pollutant whereof is carbon monoxide (CO) [2]. The problem of formation of significant amounts of CO especially rises from the process of baking graphite “green” blanks in furnaces, which are heated to

temperatures close to 1273 K, according to a specified schedule and baking rate to ensure the required quality of the electrode products. Thus, the need to convert this process into an environmentally friendly component of the electrode production requires the search for effective solutions to reduce the CO formation in the process of electrodes baking. Among the proposed steps to reduce the intensity of gas release during baking the blanks, the determinative is modernization of the baking regulations [3], use of heat exchanger-utilizer for the local finish burning of CO [4], substitution of Riedhammer multi-chamber ring furnaces for tunnel baking furnaces [5], use of catalytic methods of decontamination of CO [6, 7], *etc.* The introduction of these technical solutions at the enterprise is limited or impossible at all due to significant capital investments in the re-equipment of the production. Therefore, the solution of this issue in an economically feasible way without deterioration in the products quality is still important today.

As the study of gas release from the furnaces baking electrodes has shown [8], CO in the flue gases is formed both by the partial oxidation of the packing materials and materials of the charge mixture, as well as by the pitch degassing in the blanks. At the same time, the maximum concentration of carbon monoxide, measured directly in the chamber of the baking furnace, reached 3600 mg/dm<sup>3</sup>. It should be also noted that according to the results of previous studies, a sharp increase in CO concentration in flue gases occurs at the temperature of the blanks of 593–663 K. The CO concentration achieved in the gas mixture remains practically unchanged until emission to atmosphere. Considering that the flue gases contain oxygen at concentration of 14 % and water vapour at the concentration of 8 %, high CO content can be theoretically justified by the presence of a limiting stage of the carbon monoxide conversion into carbon dioxide. Generally, based on a slight increase in the rate constants of CO oxidation reactions for temperature, it was assumed that there is need of using catalysts for accelerating the oxidation processes. However, the use of a catalyst

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requires a clear knowledge of the source of CO formation, which in this case is limited to the general data of measuring its contents under the roof of the baking furnace chambers. Thus, there is a need to study the dependence of the amount of carbon monoxide formed on the type of carbon material used in the production of electrodes and conditions of their baking. But there is no clear data on the dependence of CO formation rates on temperature and oxygen concentration at baking the electrodes for now.

The objective of this paper is to study the processes of formation of carbon monoxide using such types of carbon material as packing materials, pitch and charge mixture for production of electrodes, depending on the

temperature regime and content of oxygen in the gas mixture, by laboratory simulation of the processes of a multi-chamber baking furnace. A prerequisite condition for the simulation process is compliance with the thermal regulations of the furnace.

## 2. Experimental

Materials used for the study were thermo-anthracite packing materials, 2.8–6 mm of fineness, granular pitch, grade B1, and charge mixture for production of the electrodes, consisting of the components presented in Table 1.

Table 1

Charge mixture formula for the electrodes manufacturing

Material	Electrically calcined anthracite			Graphite cuttings	
Granulometry	6–28	28–1	Fine powder	6–1	1–0
%	23	17	30	12	18

For simulation of gas release processes in a baking furnace a laboratory facility was created, the layout whereof is shown in Fig. 1. It includes: thermal chamber 2, consisting of ceramic tube 5 with heating element 6, which maintains preset temperature level. A sample of carbon material was placed in the thermal chamber (packing materials, pitch and charge mixture). The charge mixture and packing materials were kept in a cage of stainless steel mesh. Pitch was placed in a ceramic crucible. Temperature controller 3 connected to thermocouple 4 controlled the preset temperature level. The temperature was changed from room temperature to 1123 K discretely at different rates of the temperature rise [9]. Gas release was controlled by the change in the concentration of gas components in the flow of gases, which were slowly passed through the thermal chamber. In order to bring the composition of the gases into

proximity with the composition of flue gases in the baking furnace at production, air was supplied by compressor 9 and carbon dioxide was supplied from cylinder 10 to the thermal chamber. At the same time, the oxygen content in the mixture of gases was maintained at the level of 13–15 %, 15–16 % and 17–18 %. Of course, the content of carbon dioxide exceeded its content in the flue gases. However, formation of carbon monoxide mainly depended on the oxygen content and its interaction with carbon materials. The composition of the gases at the outlet was monitored using gas analyzer 1. The gases (air and CO<sub>2</sub>) were monitored and maintained at preset level using rotameters 7. Rotameters were calibrated for gas consumption in dm<sup>3</sup>/min. At this stage of the study, the temperature was raised and maintained at certain level for 7 h. In the course of the research, the initial and final mass of carbon material was controlled.

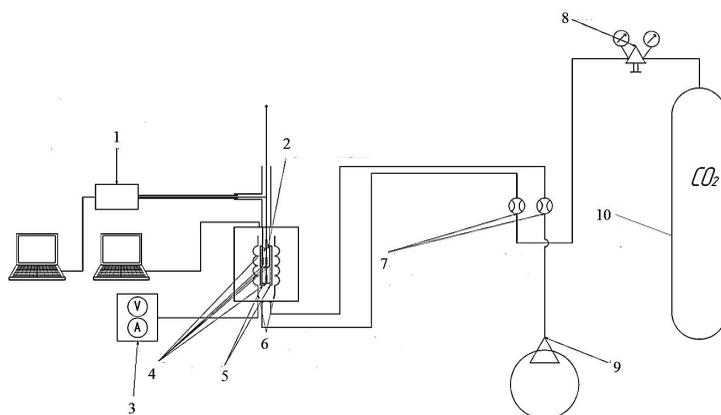


Fig. 1. Layout of the laboratory facility for simulation of gas release processes in a baking furnace: 1 – gas analyzer VARIO PLUS industrial analyzer MRU air fair; 2 – heat chamber; 3 – TENSE PC-96 temperature controller; 4 – thermocouples; 5 – ceramic wall of the thermal chamber; 6 – heating element; 7 – RF-RFM LGFI.407142.002 EF rotameters; 8 – DONMET BKO-50 DM reducer; 9 – compressor; 10 – carbon dioxide cylinder

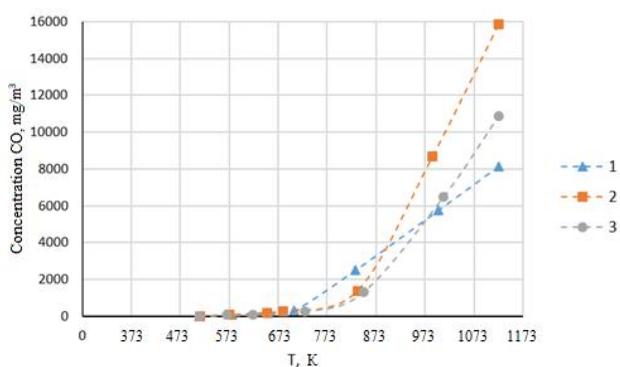
To analyze the results obtained, the specific amount of carbon monoxide formed per 1 g of loading (mg/g·h) was calculated by the formula:

$$Q_{CO} = \frac{C \cdot V}{M} \quad (1)$$

where  $C$  is the concentration of CO, mg/m<sup>3</sup>;  $V$  is a gas mixture flow rate, m<sup>3</sup>/h;  $M$  is a weight of loaded carbon material, g.

### 3. Results and Discussion

When determining the influence of temperature, concentration of oxygen in the gas phase and the packing materials as a kind of carbon material on the composition of gas emissions, about 445 g of the packing materials were loaded in the thermal chamber of the laboratory facility and the gas mixture was passed through it with a flow rate of 131.4 dm<sup>3</sup>/h at oxygen concentration of 13–15 %; with flow rate of 98.4 dm<sup>3</sup>/h at oxygen concentration of 15–16 % and with flow rate of 60 dm<sup>3</sup>/h at oxygen concentration of 17–18 %. The changes in the concentration of carbon monoxide in the gas mixture, which passed through the heat chamber are represented in Fig. 2.

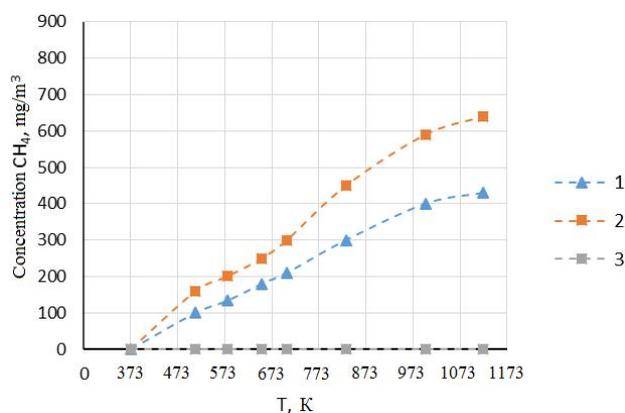


**Fig. 2.** Changes in CO concentration with rising temperature in gas mixture passed through thermal chamber loaded with packing materials ( $M = 431.4\text{--}445$  g) at the gas consumption of 131.4 dm<sup>3</sup>/h (1) ( $C_{O_2} = 13\text{--}15\%$ ); 98.4 dm<sup>3</sup>/h (2) ( $C_{O_2} = 15\text{--}16\%$ ) and 60.0 dm<sup>3</sup>/h (3) ( $C_{O_2} = 17\text{--}18\%$ )

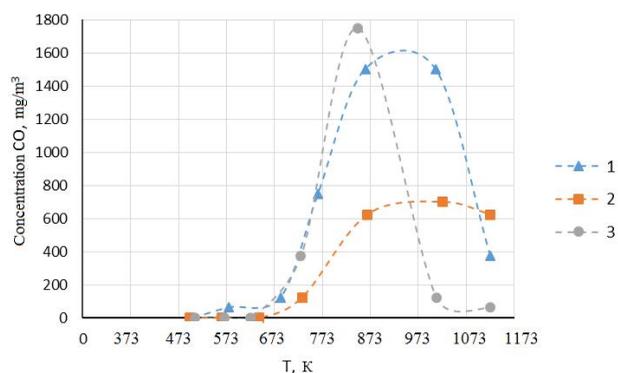
In this case, it should be noted that in the temperature range of 373–673 K, the CO concentration in the gas mixture was insignificant and only at temperatures of 523–673 K it gradually increased to 125–250 mg/m<sup>3</sup> regardless of oxygen concentration in the gas mixture. Further, up to temperature of 926 K, CO concentration grew faster at oxygen concentration of 13–15 %, and at higher temperatures – at oxygen concentration of 15–16 %. The maximum concentration of CO reached 13025–18625 mg/m<sup>3</sup> at oxygen concentration of 15–16 %.

In addition to measuring the content of carbon monoxide at heating the packing materials, the concentration of methane was monitored. As can be seen from Fig. 3, the concentration of methane increases with rising temperature. Concentrations of methane at all concentrations of oxygen used are close to each other and reach 450–650 mg/m<sup>3</sup> at the maximum temperature. There is practically no hydrogen in the gas mixture at heating the packing materials. In some cases, trace amounts of hydrogen were detected.

The loss in mass of the packing materials reached 11–15 g per 431–445 g of the initial material and reached 3.311, 2.295 and 1.947 % at oxygen concentration in the gas mixture of 13–15, 15–16 and 17–18 %, respectively. As can be seen from the results of the research, with the increase in the concentration of oxygen in the gas mixture, the loss in mass of the packing materials is minimal.



**Fig. 3.** Change in methane concentration with rising temperature in the gas mixture passed through thermal chamber loaded with packing materials ( $M = 430\text{--}455$  g) at the gas consumption of 131.4 dm<sup>3</sup>/h (1) ( $C_{O_2} = 13\text{--}15\%$ ); 98.4 dm<sup>3</sup>/h (2) ( $C_{O_2} = 15\text{--}16\%$ ) and 60.0 dm<sup>3</sup>/h (3) ( $C_{O_2} = 17\text{--}18\%$ )

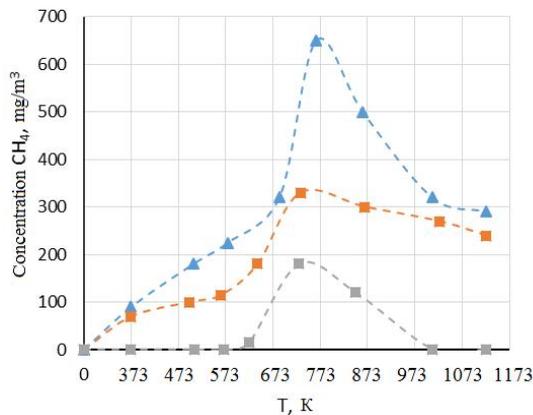


**Fig. 4.** Dependence of the carbon monoxide concentration on the temperature in the gas mixture after passing through the thermal chamber loaded with the granular pitch ( $M = 5$  g) at the gas consumption via 131.4 dm<sup>3</sup>/h (1) ( $C_{O_2} = 13\text{--}15\%$ ); 98.4 dm<sup>3</sup>/h (2) ( $C_{O_2} = 15\text{--}16\%$ ) and 60.0 dm<sup>3</sup>/h (3) ( $C_{O_2} = 17\text{--}18\%$ )

A slightly different picture was observed at heating 5 g sample weight of pitch, which is used for production of electrode mass, in heat chamber, at passing gas mixture with the consumption of 131.4, 98.4 and 60.0 dm<sup>3</sup>/h at oxygen concentrations of 13–15, 15–16 and 17–18 %, respectively (Fig. 4).

It is obvious that in this case, in addition to the concentration of oxygen, oxidation of the pitch is affected by the quantity of oxygen passed, which significantly decreases from the first to the third case. In this case, a significant release of carbon monoxide is observed already at 614 K at the maximum consumption of the gas mixture. At the minimum consumption of the gas mixture, the release of carbon monoxide began at 689 K, and at the gas consumption of 98.4 dm<sup>3</sup>/h, the gas release began at 739 K. But, with the temperature rising above 914–989 K, the CO concentration in the gas mixture decreases.

Sufficiently high concentrations of methane were achieved when the pitch was heated in the thermal chamber. Its main mass released at temperatures of 652–938 K (Fig. 5). Hydrogen release was not observed.

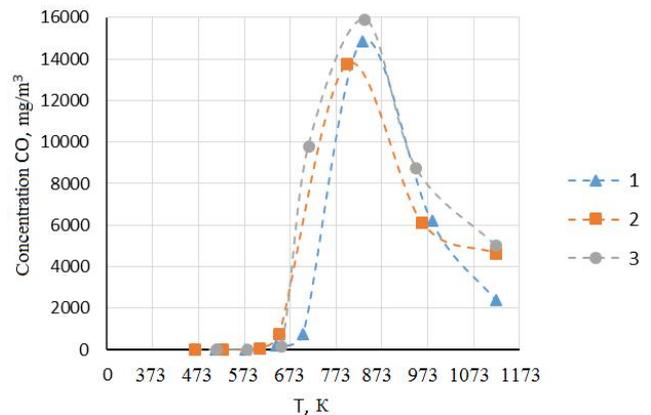


**Fig. 5.** Dependence of methane concentration on the temperature in the gas mixture after passing through the thermal chamber loaded with the granular pitch ( $M = 5$  g) at the gas consumption of 131.4 dm<sup>3</sup>/h (1) ( $C_{O_2} = 13\text{--}15\%$ ); 98.4 dm<sup>3</sup>/h (2) ( $C_{O_2} = 15\text{--}16\%$ ) and 60.0 dm<sup>3</sup>/h (3) ( $C_{O_2} = 17\text{--}18\%$ )

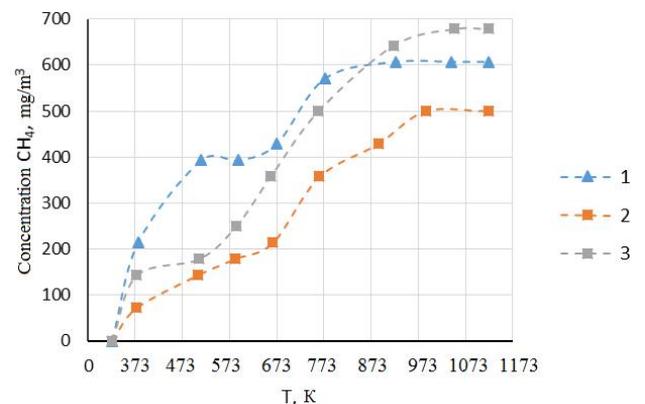
The next stage of the research was to determine the dependence of the composition of gas emissions on the temperature and oxygen content when using the charge mixture as a carbon material for manufacturing the electrodes with a gas mixture consumption of 135.2, 131.4 and 98.4 dm<sup>3</sup>/h at oxygen concentrations of 13–15, 15–16 and 17–18 %, respectively. The sample weight of the charge mixture in this case was 450 g. As a result of the research it was found that a significant formation of carbon monoxide was observed at the temperatures above 693–723 K and up to 873–953 K (Fig. 6).

With regard to the formation of methane, as in the case of using the packing materials as a loading, the concentration of methane increases with the heating temperature of the charge mixture and changes at the maximum temperature in the range of 500–680 mg/m<sup>3</sup> (Fig. 7).

Hydrogen in the gas mixture releases in small amounts when the charge mixture is heated to the temperatures of 873–973 K. Its concentration varies in the range of 4–67 mg/m<sup>3</sup>.



**Fig. 6.** Change in the carbon monoxide concentration with rising temperature after passing through the thermal chamber loaded with the charge mixture ( $M = 450$  g) at the gas consumption of 135.2 dm<sup>3</sup>/h (1) ( $C_{O_2} = 13\text{--}15\%$ ); 131.4 dm<sup>3</sup>/h (2) ( $C_{O_2} = 15\text{--}16\%$ ) and 98.4 dm<sup>3</sup>/h (3) ( $C_{O_2} = 17\text{--}18\%$ )



**Fig. 7.** Change in methane concentration with rising temperature after passing through the thermal chamber loaded with the charge mixture ( $m = 450$  g) at the gas consumption of 135.2 dm<sup>3</sup>/h (1) ( $C_{O_2} = 13\text{--}15\%$ ); 131.4 dm<sup>3</sup>/h (2) ( $C_{O_2} = 15\text{--}16\%$ ) and 98.4 dm<sup>3</sup>/h (3) ( $C_{O_2} = 17\text{--}18\%$ )

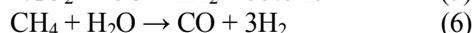
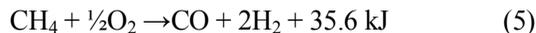
It is known that carbon monoxide, along with carbon dioxide, is formed by the interaction of solid carbon with oxygen [2]:



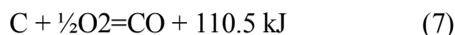
Reaction of decomposition of the complex C<sub>3</sub>O<sub>4</sub> in the presence of oxygen follows the scheme:



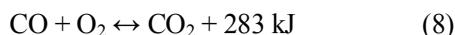
Formation of carbon monoxide due to the interaction of methane with oxygen or water vapour is also possible:



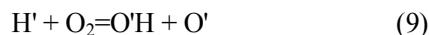
And oxidation of graphite with oxygen is possible, too:



Here, at high temperatures in the presence of oxygen, carbon monoxide can be oxidized up to carbon dioxide:



The reaction takes place in the presence of water vapour traces. In this case, transformation of CO into CO<sub>2</sub> passes mainly through the following intermediate reactions:



It should be noted that in the temperature range of 573–873 K the reaction rate constant (Eq. (10)) is quite significant and varies little in the range of (1.51–2.57)·10<sup>12</sup> cm<sup>3</sup>/mol·s. Whereas the rate-limiting step of formation of O'H radicals in the reaction (9) depends to a large extent on the temperature, and the reaction rate constant increases in the same temperature range from 7.28·10<sup>-1</sup> cm<sup>3</sup>/mol·s to 6.89·10<sup>4</sup> cm<sup>3</sup>/mol·s, that is, almost 5 orders of magnitude. Obviously, the entire process of oxidation of carbon monoxide significantly accelerates with rising temperature.

Thus, concentration of CO in the gas mixture in all cases is the result of a dynamic equilibrium between Eqs. (2)-(4) and, possibly, Eqs. (5)-(7), which leads to the formation of carbon monoxide, and Eq. (8), which goes according to the scheme: Eq. (9) – Eq. (10), and provides oxidation of carbon monoxide up to carbon dioxide.

Summarizing the results of the experimental studies, we can say that the temperature range, in which CO forms during baking the carbon materials, is significantly different from the previously published data. It was believed that the bulk of carbon monoxide is formed at temperatures of 593–663 K. And this was the main reason that prevented the creation of conditions for the oxidation of CO in the technological process. From the data obtained, it is evident that the carbon monoxide is actually formed at temperatures higher than 673 K, which makes it possible to oxidize CO in the presence of catalysts up to CO<sub>2</sub> and significantly reduce carbon monoxide emissions. If we compare the contribution of the components of the electrodes and the packing materials in the formation of CO, then it is quite difficult to evaluate these processes only according to the graphs (Figs. 2-7).

The fact is that supply of the gases to the heat chamber was adjusted to achieve a given concentration of oxygen and the consumption of the gases was different at different oxygen concentrations. In this case, the amount of formed CO is determined not only by its concentration in the gas mixture, but is calculated as the gas consumption for the concentration during the degassing period. Therefore, practical interest is the calculation of the value of the amount of generated gas for a certain time, reduced to a mass unit of carbon material. Just this index is calculated by Eq. (1) and is shown in Tables 2-4.

Table 2

**Dependence of the carbon monoxide amount formed from 1 g of the packing materials on the packing materials mass and temperature at the gas mixture consumption of 131.4 (I), 98.4 (II) and 60.0 (III) dm<sup>3</sup>/h**

t, h	T <sub>av</sub> , K			[CO <sub>av</sub> ], mg/m <sup>3</sup>			M, g (ΔM, %)			Q <sub>CO</sub> , mg/g·h			
	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	
1	511	513	512	0	0	0	445.0	431.4	431.4	0.0	0.0	0.0	
2	578	574	569	63	63	63	–	–	–	0.0186	0.0139	0.0087	
3	652	650	622	188	188	63	–	–	–	0.0553	0.0417	0.0087	
4	705	683	728	313	250	250	–	–	–	0.2480	0.0580	0.0377	
5	831	835	847	2813	1375	1313	–	–	–	1.2290	0.3130	0.1855	
6	1000	988	1011	4250	8688	6500	–	–	–	1.2539	1.9796	0.9040	
7	1123	1123	1123	8125	15875	10875	430 (3.371)	421.5 (2.295)	423 (1.947)	2.4000	3.6207	1.5126	
										Q <sub>CO</sub> <sup>av</sup>	0.7488	0.8610	0.3796

Thus, when using the packing materials as a carbon material at temperatures up to 673 K, the specific amount of carbon monoxide produced per 1 g of the loading reaches 0.0553 mg/g·h for the gas mixture containing 13–15 % of oxygen, 0.0417 mg/g·h at oxygen concentration of 15–16 % and only 0.0087 mg/g·h at oxygen concentration of 17–18 %. In all cases, the index increases with rising temperature and at 923–1073 K it increases to 1.2539, 1.9796 and 0.9040 mg/g·h with increasing oxygen content in the gas mixture from 13–15 % to 17–18 %; and at a temperature of 1123 K this indicator is respectively 2.400, 3.6207 and 1.5126 mg/g·h. It is obvious that with the increase in the oxygen concentration up to 15–16 %, the amount of the formed CO increases with rising temperature. In general, for the whole time of heating, the average index of the amount of CO formed is the highest for oxygen concentrations of 15–16 % and reaches  $Q_{CO}^{av}=0.8610$  mg/g·h. It is interesting to note that the

specific amount of carbon monoxide formed per 1 g of carbon load, calculated for the baking furnace at the production, for the entire production cycle of the blanks processing amounts to 0.12297 mg/g·h. Perhaps this is due to the fact that flue gases contain up to 8 % of water vapour, which accelerates the reaction of CO oxidation in the reactions given in the literature [9], which generally ensures a reduction in its concentration in the gas mixture.

In this case, the bulk of CO releases in the temperature range of 773–1123 K with the increase in the amount of the formed carbon monoxide with rising temperature.

Increasing the concentration of methane with rising temperature can be associated with the presence of a certain amount of hydrocarbons in the mass of the packing materials, which are released with rising temperature. It is difficult to explain the formation of methane by pyrolysis at temperatures from 373 to 1123 K.

Table 3

**Dependence of the carbon monoxide amount formed from 1 g of the heated granular pitch on the temperature and heating time at the gas mixture consumption of 131.4 (I), 98.4 (II) and 60.0 (III) dm<sup>3</sup>/h**

t, h	T <sub>av</sub> , K			[CO <sub>av</sub> ], mg/m <sup>3</sup>			M, g (ΔM, %)			Q <sub>CO</sub> , mg/g·h		
	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )
1	504	496	507	0	0	0	5.0	5.0	5.0	0.0	0.0	0.0
2	577	563	569	63	0	0	–	–	–	1.6425	0.0	0.0
3	686	640	622	63	0	0	–	–	–	1.6425	0.0	0.0
4	764	731	728	438	63	250	–	–	–	11.4975	1.6425	6.5700
5	862	867	847	1125	375	970	–	–	–	29.5650	9.3550	25.4900
6	1009	1024	1011	938	1163	438	–	–	–	24.6375	17.4105	11.4900
7	1123	1123	1123	375	375	63	2.2 (56)	2.5 (50)	2.5 (50)	8.8550	9.855	1.6425
										Q <sub>CO</sub> <sup>av</sup> 11.120	4.2705	6.4561

Table 4

**Dependence of the carbon monoxide amount formed from 1 g of the heated charge mixture on the temperature and heating time at the gas mixture consumption of 131.4 (I), 98.4 (II) and 60.0 (III) dm<sup>3</sup>/h**

t, h	T <sub>av</sub> , K			[CO <sub>av</sub> ], mg/m <sup>3</sup>			M, g (ΔM, %)			Q <sub>CO</sub> , mg/g·h		
	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )	I (13-15 % O <sub>2</sub> )	II (15-16 % O <sub>2</sub> )	III (17-18 % O <sub>2</sub> )
1	510	465	513	0	0	0	450	450	450	0.0	0.0	0.0
2	575	525	580	0	0	0	–	–	–	0.0	0.0	0.0
3	643	606	653	188	63	125	–	–	–	0.0563	0.0188	0.0376
4	701	649	713	438	438	5025	–	–	–	0.1314	0.1314	1.5097
5	830	797	836	7625	7250	12850	–	–	–	2.2908	2.1170	3.8607
6	982	961	948	6188	6125	8750	–	–	–	1.8600	1.8402	2.6289
7	1123	1123	1123	2375	4625	5000	409 (9.1)	411 (8.67)	405 (10)	0.7139	1.3895	1.5022
										Q <sub>CO</sub> <sup>av</sup> 0.7217	0.7852	1.3627

Reducing the mass of the packing materials with increasing oxygen concentration can be associated with a decrease in the consumption of the gas-air mixture, which in general led to a decrease in the amount of the oxygen passed. Thus, at oxygen concentration of 13–15 %, the gas consumption was 131.4 dm<sup>3</sup>/h, or 919.8 dm<sup>3</sup> for 7 h, where 14 % or 128.772 dm<sup>3</sup> is oxygen. At oxygen concentration of 15–16 %, the gas mixture consumption was 98.4 dm<sup>3</sup>/h, or totally 688.8 dm<sup>3</sup>, including 106.76 dm<sup>3</sup> of oxygen. At the consumption of the gas mixture of 60 dm<sup>3</sup>/h, the amount of the oxygen passed is only 73.5 dm<sup>3</sup>. Just this explains the reduction in the amount of carbon monoxide formed at reducing the consumption of the gas mixture.

As for the pitch, the choice of its mass was determined by the experimental conditions, its melting temperature and high content of resinous substances [10], which prevented the normal operation of the gas analyzer. In the course of the studies with the pitch, a sharp increase in the concentration of CO at the temperature of 614 K at the maximum consumption of the gas mixture can be explained by the content in this load of resinous substances that begin to oxidize first. Further decrease in the concentration of CO at the temperatures above 914–989 K is associated both with a higher rate of oxidation of organic substances of the pitch and with a small weight of the used pitch sample (only 5 g), which is almost 100 times less than the heated packing materials. Therefore, the reduction of CO concentration when heating above the temperature of 914 K in the gas mixture can be argued by the exhaustion of the bulk of volatile substances of the pitch. After all, the weight loss of the pitch (Table 3) in this case was 56.0, 50.0 and 50.0 % at the gas consumption of 131.4, 98.4 and 60.0 dm<sup>3</sup>/h, respectively.

Undoubtedly, the pitch contains volatile substances that are easily oxidized with the formation of carbon monoxide. As can be seen from Table 3, at different gas consumptions, the specific rate of oxidation of organic substances in terms of 1 g of organic mass per hour reaches 17.4105, 25.490 and even 29.5650 mg. This is an order of magnitude more than in the case of the packing materials. In general, the amount of the formed carbon monoxide directly depends on the amount of oxygen supplied, which in this case depends more on the gas consumption than on the oxygen concentration.

High concentrations of CO formed in the studies with the charge mixture can undoubtedly be justified by the fact that the crushed charge mixture has a developed surface, which specific area of contact with air is

significant. The proof of this fact is a decrease of the charge mixture mass by 9 %. However, taking into account that  $Q_{CO}$  value reached 0.113140–3.8607 mg/g·h (Table 4), it can be concluded that the specific amount of carbon monoxide from the charge mixture prevails that from the packing materials, but is inferior to the pitch.

Since the experimental temperature was lower than the pyrolysis temperature, and there was nearly no water vapour in the gas mixture, then there was some reason to wait for a high concentration of methane, which was confirmed experimentally.

Taking into account the given results, which show that carbon monoxide is formed at sufficiently high temperatures, it is quite promising to use water vapour and other catalysts to significantly reduce its amount.

## 4. Conclusions

Thermal oxidation processes of carbon components (packing materials, pitch, charge mixture) were studied at passing the gas mixture with oxygen concentration of 13–15, 15–16 and 17–18 % within the temperature range up to 1123 K. It has been shown that when heating the packing materials, the bulk of carbon monoxide is formed at 773–1123 K, when heating the pitch – at the temperatures of 614–989 K, when heating the charge mixture – at 693–953 K. It has been determined that due to the presence of large amount of volatile substances, the pitch allows the highest rate of carbon oxidation with the release of CO and significantly exceeds this index for the packing materials and the charge mixture. The specific amount of carbon monoxide formed per gram of carbon load per hour in the range up to 1123 K has been calculated. It is established that this index depends on the type of carbon material, gas mixture consumption, oxygen concentration and temperature regime.

## References

- [1] Kutuzov S., Buriak V., Churilin V. *et al.*: IV Int. Congress “Non-Ferrous Metals”. Russia, Krasnoyarsk 2012, 943.
- [2] Panov Ye., Shylovykh I., Ivanenko E. *et al.*: East.-Eur. J. Enterpr. Technol., 2012, **58**, 15.
- [3] Karvatskyi A., Leleka S., Pulinets I. *et al.*: East.-Eur. J. Enterpr. Technol., 2011, **54**, 42.
- [4] Karvatskyi A., Shylovykh I., Krutous L. *et al.*: East.-Eur. J. Enterpr. Technol., 2013, **62**, 38.
- [5] Bogacki M., Oleniacz R., Mazur M. *et al.*: Environ. Protect. Eng., 2012, **38**, 15.

- [6] Rattan G., Kumar M.: Chem. Chem. Technol., 2014, **8**, 249.  
<https://doi.org/10.23939/chcht08.03.249>
- [7] Kostrobii P., Ryzha I.: Chem. Chem. Technol., 2018, **12**, 451.  
<https://doi.org/10.23939/chcht12.04.451>
- [8] Gomelya N., Ivanenko E.: The IX International Youth Science Environmental Forum "Ecobaltica-2012". Russia, St-Petersburg 2012, 89.
- [9] Fokin V., Malakhov A., Malakhov S. et al.: Tsvetnye Metally, 2002, **4**, 48.
- [10] Mazur M., Oleniacz R., Bogacki M. et al.: Emission of polycyclic aromatic hydrocarbons (PAHs) during the production of carbon and graphite electrodes [in:] Pawłowski L., Dudzińska M., Pawłowski A. (Eds.), The 3<sup>rd</sup> Congress of Environmental Engineering, v.3. CRC Press/Balkema, Taylor & Francis Group, Leiden-London 2010, 59-66.

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### **ВИЗНАЧЕННЯ УМОВ ОКИСНЕННЯ КАРБОНОВИХ МАТЕРІАЛІВ З УТВОРЕННЯМ ОКСИДУ КАРБОНУ ЗА ВИСОКИХ ТЕМПЕРАТУР**

**Анотація.** Експериментальним шляхом досліджено вплив типу карбонового матеріалу, температури та концентрації кисню в газовій суміші на процеси утворення оксиду карбону при виробництві електродів шляхом графітації в печах обпалу. Розраховано питому кількість утвореного газу за певний час, приведений до одиниці маси карбонового завантаження на прикладі пересипки, пеку та шихти промислового застосування. Показано, що пек забезпечує найвищу швидкість окиснення карбону з виділенням CO і суттєво за цим показником переважає пересипку і шихту.

**Ключові слова:** оксид карбону, газовиділення, пересипка, пек, шихта.