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Obtaining Carbon Sorbent from "Euromet" LLP Coke Breeze

The paper presents the results of a physicochemical analysis of a coke sample from "Euromet" LLP (Karaganda, Kazakhstan), which is carbonaceous material with high carbon content, the absence of volatiles matter and low ash content. Carbon sorbents were obtained from low-value fractions of carbonized materials of the Karaganda region coke production by the steam-gas activation. An important advantage of using screenings (coke breeze class 0–10 mm, which was crushed and fractionated to a particle size of 2–5 mm.) is the exclusion of the energy-intensive carbonization stage from the technological process, which releases a large amount of environmentally harmful substances (phenols, cresols, gaseous emissions etc.). The optimal conditions for obtaining a sorbent from low-value coke breeze are a temperature of 850 °C and an activation time of 2 hours. The physicochemical properties (ash content, bulk density, sorption capacity for iodine and methylene blue, total pore volume for water) of carbon sorbents were determined.

Keywords: coke breeze, coals, sorbents, activation, sorption capacity, ash content, burning, bulk density.

Introduction

Environmental pollution by the oil, coal and metallurgical industries is one of the reasons for the severe environmental consequences and the negative impact on human health. Industrial enterprises of the republic discharge insufficiently treated wastewater into water sources. In addition, the water resources of Kazakhstan are polluted by household and agricultural waste. Currently, there is a tightening of requirements for the standards of discharges and emissions from production and economic activities. The high sorption activity of active carbons determines a wide range of their application, including the possibility of using them for purification from pollutants. The use of this product in the world is steadily growing with the development of industrial production of activated carbon. So in Western Europe, activated carbons are used as the main material for wastewater treatment and gas emissions, in accordance with current environmental regulations. The areas of application of carbon sorbents are inextricably linked both with the knowledge of their sorption properties in relation to various pollutants and with their physicochemical properties.

Also, in addition to activation gases, various chemical reagents are used at the stages of carbonization and preparation of raw materials. Different vegetable raw materials such as wood, nutshell, remains of stems and seed coats [1, 2], as well as processed carbon-containing substances such as plastics, rubber, oil, coal [3], are used as raw materials for the production of carbon sorbents.

A large amount of low-value fraction (screenings) remains during the preparation of products for sale at enterprises producing coke using different technologies. The use of this material will make it possible to obtain highly effective carbon sorbents for the public utilities, chemical, oil, gas, and mining industries with certain processing.

The advantage of using screenings is the exclusion from the technological process of the energy-consuming carbonization stage, which releases a large amount of environmentally harmful substances (phenols, cresols, gaseous emissions, etc.). The ash content, bulk density, and average particle diameter are linearly related to the degree of burning during activation. However, it is necessary to consider the specifics of the norms of sorbents for various purposes. Most of them require such parameters as grain size, total pore volume in water, mass fraction of moisture, bulk density, and abrasion resistance.

The grade of coal should be considered when activating hard coal [4]. Bituminous coals with a high tar and volatiles content, sinter or swell when heated, and therefore require pre-treatment. Another activation process [5] for coals of the “bituminous C” class with a high content of volatiles and moisture is carried out on grained raw materials, which are first subjected to drying and preliminary oxidation at 150–215 °C. Anthracite, which contains significantly fewer volatiles, can be activated immediately under appropriate conditions. To do this, anthracite is first crushed, the finely dispersed powder is briquetted with a binder, re-crushed, and after sieving into fractions, it is subjected to carbonization and activation. Direct activation of granular anthracite is difficult to implement and requires a long time in conventional rotary kilns. Low-grade bituminous coals (fat coals) with a relatively high volatile content, which coke poorly and produce grains of low strength, can also be used to produce activated carbons if crushed and washed with dilute mineral acid (hydrochloric, sulfuric, or phosphoric). Then after drying, the grains are crushed, the powders are granulated with binders, carbonized, and activated [6]. It is possible to activate the products of various stages of molded coke production with gas [7]. Coal-based activated carbons are not produced in Central Kazakhstan. The quality of the finished activated carbon depends on the correct set of technological parameters for the preparation of the initial material, activation modes. At present, the production of activated carbon is promising due to the possibility of obtaining materials with a developed structure for solving various problems, including protection of the biosphere from anthropogenic impact, in nuclear power engineering, and in modern devices for energy storage and transmission. At the same time, the scale of the use of nanoporous carbon is limited by the rather high cost of materials. This leads to the search for new methods for obtaining carbon materials with the required set of properties from cheap types of raw materials, which is an urgent task for science and industry [8]. For example, the cost of adsorbents is 450 times higher than the cost of raw materials with a huge sales market, and the total world production of porous carbon materials is currently about 1 million tons/year [9, 10].

With the high rates of development of heavy and light industry, motor transport entail an increasing load on surface waters [11, 12]. The quality of some natural water sources is unacceptable for their use not only for food purposes but also for technical needs. Hazardous organic substances, heavy metals, and radionuclides may be present in water bodies. Such water is of poor quality and certainly needs to be cleaned. The use of powdered activated carbon to capture mercury is actual for the Karaganda region (Nura river). In [13], the porous carbon materials obtained from the coal of the Maikuben basin, the Shoptykol deposit (Kazakhstan), with a specific surface area (348.99 m^2/g) were tested to treat waste sewage water taken from the SUC “Astana Water Canal” (Nur-Sultan, Kazakhstan). Active carbon is the only type of sorbent that has a high adsorption capacity when extracting toxic organic contaminants from water. All drinking water supply and deep (below maximum permissible concentrations) wastewater treatment are based on the use of powdered and granular activated carbons. For these purposes, 30 % of the global production of activated carbon is spent.

According to experts, it is necessary to produce at least 0.5 kg of activated carbon per capita per year for an environmentally safe and healthy lifestyle of people. This rate is undoubtedly higher in regions with high environmental pollution (for the Republic of Kazakhstan, at least 7.5 thousand tons per year). The main areas of use of carbon sorbents are associated with technological processes of adsorption purification, separation, isolation and concentration in gaseous and liquid media. Volatile organic compounds are the main pollutants of various industrial installations [14–16]. Some volatile organic compounds, such as toluene vapor released into the atmosphere as a result of their use, lead to serious environmental problems, therefore the concentration of which should be reduced to low levels in the environment [17–19]. Adsorption by activated carbon is one of the most common processes for the removal of volatile organic compounds, especially toluene from gaseous media [20, 21] and phenol from chemical wastewater [22].

In the world market, the main producer of activated carbon in the United States accounts for 34 % of the annual production of this product category. Europe, with a share of 24 %, ranks second in terms of activated carbon production. Kazakhstan accounts for only 0.7 % of the global activated carbon production, while Russia accounts for 2 % (Fig. 1).

Demand for activated carbon is growing due to its usefulness as a carbonaceous material in the fight against pollution. As a result, the cost of activated carbon rises depending on the application. The research of scientists is carried out in the direction of searching for a raw material base for the production of activated carbon for its low cost, availability, high carbon content and low ash content. The cost of coke breeze in the Republic of Kazakhstan is 60–90 thousand tenge/t. Activated coals based on coal, depending on the brand, cost from 800 thousand tenge/t. This indicates the need to organize their own production facilities for their production.

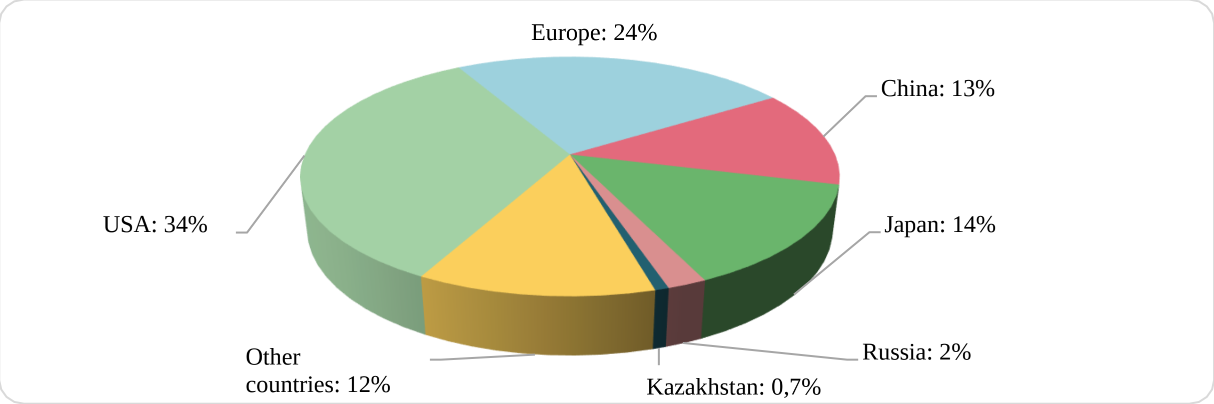

Figure 1. Share distribution of the annual production of activated carbon by country

This paper aims to determine the possibility of using a low-value fraction of the coke production of "Euromet" LLP (Karaganda, Kazakhstan) as a feedstock for the production of carbon sorbents.

Experimental

The object of the study was the coke breeze of the 0–10 mm class, which was subjected to crushing and fractionation to a particle size of 2–5 mm. Activation of coke from Karaganda coal was carried out on a laboratory unit based on a high-temperature tube furnace with maximum heating of 1100 °C and a quartz reactor. The amount of steam produced is controlled by the steam generator heater.

To work out the optimal conditions for obtaining active carbon, the feedstock was dried at a temperature of 120 °C for 3 hours, then 150–180 grams were loaded into the reactor, and, after connecting all systems, heating of the furnace was turned on. At 550–700 °C, the components absorbed during the quenching of coke are removed. At a temperature of 800 °C, steam is supplied to the reactor from the steam generator. After reaching the temperature of the set mode, activation continued for 1–2 hours, while the volatile substances condensed in the receiving flask cooled with water, and the resulting gases were removed carbon of the studied coke. The temperature at which the process of interaction of water vapor with the carbon of the coke under study begins was determined using GLC analysis on the "Crystallux 4000 M" (Research and Production Association "MetaKhrom", Russia) chromatograph. The relative average quadratic change in the output signal for a detector of an accident chromatograph "Crystallux 4000 M" is no more than 2 %. The accuracy of measurement results was estimated using a standard deviation (δ) according to [23, 24]. The correction factor T is 2.7760 for a 95 % level of reliability (trust probability) of a series of 5 measurements. To calculate the critical value F, the MS Excel program was used, where regression equations were obtained for the indicators: burnt, ash content, bulk density, sorption capacity for iodine and methylene blue, total pore volume for water, and structural strength.

250–300 g of dry coke with a grain size of 2–5 mm was placed in the reactor, the steam generator was turned on and, after boiling water, the furnace was turned on. Activation temperatures were 750, 800 and 850 °C. The set time for heating the oven to the set temperature was 10 minutes; the duration of activation was 60 and 120 minutes. At the end of the activation time, the heating and steam supply were stopped and, after the reactor cooled down, the yield of the obtained sorbent, the degree of burning, and bulk density were determined. The analysis of the obtained products was carried out according to the procedures [25].

Results and Discussion

Table 1 shows the main indicators of coke breeze from “Euromet” LLP.

Table 1

Indicators of physico-chemical analysis of coke breeze from “Euromet” LLP

The name of indicators	UM	Medium
Mass fraction of carbon, C^d	%	upto 84
Ash content on dry state, A^d	%	upto 18.0
Mass fraction of total moisture	%	upto 5.0
Structural strength of dry coke	%	78–82

According to the given parameters, coke breeze from the coals of “Euromet” LLP can be used as a raw material for the production of carbon sorbents. Table 2 shows the data and test results from the laboratory of “Coal Research and Development Center” LLP, characterizing the degree of coke activation for the “Euromet” LLP coals in terms of its burning, ash content, bulk density, total pore volume in water, iodine and methylene blue activity.

Tables 2 and 3 demonstrate the boundaries of the confidence interval (±Δ) of the measured characteristics of the “Euromet” LLP sorbent, determined considering the Student’s criterion.

Table 2

Conditions for obtaining and characteristics of the “Euromet” LLP sorbent

The mode for sorbent obtaining		Combustion loss, %	Ash content, %	Bulk density, g/dm^3
T, °C	duration of activation, hour			
750	2	20.71±0.63	23.33±0.49	330±4.5
800	1	20.90±0.32	24.04±0.50	333±4.3
800	2	22.00±0.42	24.75±0.51	335±4.2
850	1	23.11±0.49	25.78±0.54	338±3.9
850	2	34.76±0.49	29.82±0.45	339±3.8

Table 3

Conditions for obtaining and characteristics of the “Euromet” LLP sorbent

The mode for sorbent obtaining		Sorption capacity		Total pore volume by water, cm^3/g	Structural strength, %
T, °C	duration of activation, hour	for iodine, %	Methylene blue, mg/g		
750	2	22.50±0.41	55±0.9	0.5258±0.0102	9.50±0.115
800	1	23.05±0.53	50±1.1	0.3533±0.0089	9.40±0.207
800	2	24.20±0.58	20±1.2	0.5975±0.0081	9.70±0.158
850	1	25.50±0.60	100±1.4154	0.5711±0.0167	9.60±0.207
850	2	27.94*±0.63	130±1.4	0.7501*±0.0086	9.80*±0.158

For the combustion loss index, the regression equation with different variations in the temperature and time modes of coal processing (Table 2) is described by the following equation:

$$Y\,(\text{combustion loss}) = 0.1244T + 9.32t - 92.34. \qquad (1)$$

The coefficient of determination R-square is 0.83, the critical value F is 5.03, which is greater than the significance criterion F, equal to 0.16, which confirms the significance of the burn value from the mode of obtaining the sorbent. From equation (1), the amount of combustion loss increases linearly with increasing temperature and duration of the process.

The following equation describes the regression equation for the ash content index from the temperature and time modes of coal processing (Table 2):

$$Y\,(\text{ash content}) = 0.030902T + 3.00523t - 3.923233. \qquad (2)$$

In this case, the value of R-square is 0.15, the critical value of F is 0.18, which is less than the significance criterion F, equal to 0.84, which indicates the significance of the value of the composition of the initial coal, but does not depend on the mode of its processing. Analysis of equations (2) shows that the ash content depends on the temperature and duration of the process, due to a decrease in the mass fraction of carbon.

The regression equation linking the obtained indicators for bulk density with different variations in the temperature and time modes of coal processing (Table 2) is as follows:

$$Y\text{ (bulk density)} = 0.144T + 1.4t - 471.4. \quad (3)$$

The value of the R-square is 0.03, and the critical value F 0.03 is less than the significance criterion F 0.96, which also indicates the significance of the value of the composition of the initial coal, but does not depend on the processing mode. An analysis of equation (3) shows that the bulk density value slightly depends on the process temperature and increases with the length of the processing time.

A regression equation has been obtained that relates the obtained indicators for the iodine sorption capacity with different variations in the temperature and time modes of coal processing (Table 3):

$$Y\text{ (sorption capacity for iodine)} = 0.05332T + 1.938t - 21.652. \quad (4)$$

The R-square value is 0.96; the critical value F 25.19 is greater than the significance criterion F 0.03, which indicates the significance of the indicator from the mode of obtaining the sorbent.

A regression equation was obtained that relates the obtained indicators for the sorption capacity for methylene blue with different variations in the temperature and time modes of coal processing (Table 3):

$$Y\text{ (sorption capacity for methylene blue)} = 0.8T + 13.33t - 598.333. \quad (5)$$

The value of the R-square is 0.53, the critical value F 1.36 is greater than the significance criterion F 0.46, which indicates the significance of the indicator from the mode of obtaining the sorbent.

A regression equation has been obtained that relates the obtained indicators for the total pore volume in water with different variations in the temperature and time regimes of coal processing (Table 3):

$$Y\text{ (total pore volume by water)} = 0.002666T + 0.228907t - 1.96583. \quad (6)$$

The R-square value is 0.93, the critical value F 13.68 is greater than the significance criterion F 0.06, which indicates the significance of the indicator from the mode of obtaining the sorbent.

A regression equation has been obtained that relates the obtained indicators for structural strength with different variations in the temperature and time modes of coal processing (Table 3):

$$Y\text{ (structural strength)} = 0.0032\mathrm{T} + 0.246667t - 6.613333. \quad (7)$$

The R-square value is 0.97, the critical value F 36.5 is greater than the significance criterion F 0.02, which indicates the significance of the indicator from the mode of obtaining the sorbent.

Equations (4–7) show that the contribution of temperature to the increase in sorption capacity is significant for the indicator of sorption capacity for iodine and methylene blue, and for indicators of the total pore volume for water and structural strength, which will allow us to conclude that macropores in the sorbent increase. The value of the sorption capacity for iodine increases with increasing temperature treatment, which is consistent with theoretical ideas about the process.

Methylene blue activity indicates the AC surface formed by pores larger than 1.5 nm since the indicator molecule is relatively large. Therefore, low activity indicators for iodine, which is adsorbed on the surface of pores with a diameter significantly more than 1 nm, and methylene blue are not an indicator of the low quality of the carbon sorbent [26]. With a large specific surface, the proportion of thin pores increases, which do not fit large molecules, for example, persistent organic pollutants such as phenols, aromatic and polyaromatic hydrocarbons, etc. The results of sorption activity for iodine (MRTU No6-16-1003-67) [27] and clarifying ability for methylene blue (GOST 4453–74) [28] showed that the optimal conditions for obtaining a sorbent from coke breeze are: temperature 850 °C and activation time 2 hours. Properties of activated carbons depend on the choice of the appropriate raw material, method, and activation conditions. At the same time, a number of conditions can affect certain properties. Thus, the number and distribution of pore sizes depend, in particular, on the nature of the raw material, type and conditions of the activation process. The formation of gaseous products (H_2, CO, CO_2, CH_4) begins at a temperature of 670^0C, which indicates the presence of water vapor with the carbon of the coke breeze.

It is established that the exhaust gas contains hydrogen, carbon dioxide, methane, and carbon monoxide. When carbon interacts with water vapor or carbon dioxide, the following reactions occur simultaneously (Equations 8–10):

$$H_2O + C = CO + H_2 \qquad \Delta H = +117\ \text{kJ}; \tag{8}$$

$$2H_2O + C = CO_2 + 2H_2 \qquad \Delta H = +75\ \text{kJ}; \tag{9}$$

$$CO_2 + C = 2CO \qquad \Delta H = +159\ \text{kJ}. \tag{10}$$

Table 4 shows the composition of gases obtained during the activation of coke breeze of “Euromet” LLP.

Table 4

The composition of the gas obtained during the activation of “Euromet” LLP coke breeze

Gas	Gas outlet mode							
	T,°C	τ, min	T,°C	τ, min	T,°C	τ, min	T,°C	τ, min
	670	120	750	120	800	120	850	120
H_2	50.33		55.23		59.08		53.7	
O_2	–		–		–		–	
N_2	0.780		0.80		0.95		0.95	
CO_2	18.90		20.40		15.43		10.74	
CH_4	3.35		3.05		3.38		0.89	
CO	17.42		20.52		21.16		33.72	

Thus, a carbon sorbent with a high adsorbing property was obtained as a result of the high-temperature process of activation of the coke breeze with water vapor on the basis of “Euromet” LLP (Karaganda, Kazakhstan) coal.

Conclusions

Physical and chemical analyzes showed that a sample of coke breeze from “Euromet” LLP (Karaganda, Kazakhstan) is a highly active carbonaceous material with high carbon and low ash content. The optimal technological parameters for obtaining carbon sorbents from screenings of coke production at a laboratory facility (temperature 850 °C and activation time 2 hours) were established. The conditions for coke breeze activation and characteristics (combustion loss, ash content, bulk density, total pore volume in water, and activity in terms of iodine and methylene blue) of the obtained sorbent were determined.

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«Евромет» ЖШС кокс ұнтағынан көміртекті сорбент алу

Мақалада құрамында көміртегі жоғары, ұшпа және күлділігі төмен көміртекті материал болып табылатын «Евромет» ЖШС (Қарағанды қ.) кокс ұнтағы үлгісінің физика-химиялық талдау нәтижелері келтірілген. Көміртекті сорбенттер Қарағанды облысының коксхимия өндірісінің карбонизацияланған материалдарының аз ликвидті фракцияларынан бу-газды активтендіру әдісімен алынды. Қалдықтарды пайдаланудың маңызды артықшылығы (ұсақтауға және 2–5 мм-ге дейін фракциялауға ұшыраған 0–10 мм класты кокс ұсақтары) технологиялық процестен экологиялық зиянды заттардың көп мөлшері (фенолдар, крезолдар, газтәрізді шығарындылар және т.б.) шығарылатын карбонизацияның энергияны ысырап ететін сатысын алып тастау болып табылады. Аз ликвидті кокс ұнтағынан сорбент алудың оңтайлы шарттары активтендіру уақыты 2 сағат болған кезде 850 °C температура болып табылады. Көміртегі сорбенттерінің физика-химиялық қасиеттері (күлділігі, үйінділік тығыздығы, йод және көк метилен бойынша сорбциялық сыйымдылығы, су бойынша кеуектердің жиынтық көлемі) анықталды.

Кілт сөздер: коксты ұсақ-түйек, көмір, сорбенттер, активтендіру, сорбциялық сыйымдылық, күлділік, күйдіру, үйінділік тығыздығы.

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Получение углеродного сорбента из коксовой мелочи ТОО «Евромет»

В статье приведены результаты физико-химического анализа образца коксовой мелочи ТОО «Евромет» (г. Караганда), представляющего собой углеродистый материал с высоким содержанием углерода, отсутствием летучих веществ и низкой зольностью. Углеродные сорбенты были получены из малоликвидных фракций карбонизованных материалов коксохимического производства Карагандинской области методом парогазовой активации. Важным преимуществом использования отсевов (коксовая мелочь класса 0–10 мм, которая подвергалась дроблению и фракционированию до крупности 2–5 мм) является исключение из технологического процесса энергозатратной стадии карбонизации, при которой выделяется большое количество экологически вредных веществ (фенолы, крезолы, газообразные выбросы и т.д.). Оптимальным условием получения сорбента из малоликвидной коксовой мелочи является температура 850^{0}С при времени активации 2 ч. Определены физико-химические свойства (зольность, насыпная плотность, сорбционная емкость по йоду и по метиленовому голубому, суммарный объем пор по воде) углеродных сорбентов.

Ключевые слова: коксовая мелочь, угли, сорбенты, активация, сорбционная емкость, зольность, обгар, насыпная плотность.

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