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Environmental Issues in Kuznetsk Coal Basin. Scientific Approaches and Technologies to Reduce Environmental Pollution

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Abstract

Two major groups of environmental problems of Kuznetsk Coal Basin are discussed: firstly, the investigation of the current state of pollution level and its monitoring and secondly, pollution reduction research and technologies. Justification and development of methods, approaches, and instruments ensuring the integrated and systemic determination of pollutants that have a negative impact on the environment during coal production and conversion are given. The presence of 2,4,6-trinitrotoluene and its metabolites in environmental objects is explored in detail. The content of polycyclic aromatic hydrocarbons in various metamorphic states of coal is determined by modern analytical methods. The composition of solid wastes from coal production and conversion and the distribution of gross, mobile, and water-soluble forms of toxic elements and organic compounds therein are investigated. The issue of toxic fine coal materials formed during coal production, transportation, preparation, and conversion is considered. The composition of gaseous pollutants entering the atmospheric air from coal-beds is explored. To address pollution reduction issues, a number of catalytic processes are suggested. Among them is combustion of fuels, including low-grade coal in a fluidised catalyst bed, catalytic heat generators to reduce emissions of toxic nitrogen oxides and products of incomplete combustion. Honeycomb catalysts for abatement of nitrogen oxides emissions in flue gases of existing furnace plants are used. Key findings for a wide range of chemical reactions, such as the processing of coal-bed methane into highly valuable commercial products, oxidative coupling, autothermal reforming, and synthesis of carbon nanofibers and hydrogen, are presented. Research results regarding the formation of coal-bed methane hydrates and approaches to their utilization are briefly described. The paper provides a review of works on the development and research of carbon sorbents directly from Kuzbass coal to address environmental issues of the region itself.

Keywords: environment, monitoring, coal, toxicity, waste, polycyclic aromatic hydrocarbons, 2,4,6-trinitrotoluene, catalytically assisted combustion, monolithic honeycomb catalysts, nitrogen oxides, methane hydrates, coal-bed methane, aromatization of methane, carbon nanofibers and sorbents

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INTRODUCTION

Peculiarities of the Kuzbass environmental conditions are driven by a high technogenic load onto the territory mainly related to the functioning of enterprises of coal mining and processing, and heat and power complexes. The effect of the enterprises of these complexes on the environment in the frameworks of the entire region is hard to differentiate. Coal production and conversion enterprises of the region exert the maximum environmental load. According to the estimates of the Ministry of Natural Resources of the Russian Federation, the Kemerovo region enters the list of the most problematic subjects of the Russian Federation in part of the total environmental harm caused.

Contamination and depletion of water bodies, the imperfection of the waste management system of production and consumption, and pollution and degradation of soil and land resources refer to the number of the major environmental issues of the region. Herewith, the scenario of the long-term socio-economic development of the Kemerovo region demonstrates a further increase in the anthropogenic load on all components of the natural environment.

To ensure the environmental safety and efficient realization of environmental protection measures in coal mining regions, on the one hand, it is required to substantiate and develop methods, approaches, and instruments that guarantee integrated and systemic determination of pollutants that exert a negative influence on the environment during coal production and conversion from Kuznetsk Coal Basin. On the other hand, it is necessary to develop scientific approaches and introduce specific technologies to reduce air, water, and soil pollution.

This article gives a brief overview of research carried out in different years at the Boreskov Institute of Catalysis SB RAS (BIC SB RAS), the Institute of Coal Chemistry and Chemical Materials Science of the Federal Research Centre of Coal and Coal Chemistry SB RAS (Kemerovo) and West Siberian Test Centre (Novokuznetsk).

INTEGRATED AND SYSTEMIC DETERMINATION OF POLLUTANTS

Within substantiation and development of methods, approaches, and tools to ensure a comprehensive and systemic determination of pollutants that have a negative impact on the environment during coal production and conversion in the Kuznetsk Basin, we address the following objectives:

– scientific substantiation of the list of pollutants formed during coal production and conversion for the purpose to improve environmental monitoring, environmental safety and effective implementation of environmental measures;

– analysis and selection of the most effective methods for assessing the concentration of toxic inorganic and organic pollutants in coal and environmental facilities in operating areas of coal production and conversion enterprises;

– a comprehensive assessment of the toxicity of industrial wastes of coal mining and processing industries, including the determination of the gross content of elements, their mobile and water-soluble forms, toxic organic compounds, and waste bioassay;

– determination of interrelations between the gross content of toxic elements and their mobile forms, identification of activity series of water-soluble forms of toxic elements in coal mining and processing wastes;

– development of methods for determination of 2,4,6-trinitrotoluene (TNT) and its metabolites in environmental objects and assessment of their concentration levels in the area of influence of mining enterprises;

– exploration of the distribution of polycyclic aromatic hydrocarbons (PAH) in environmental objects in the area of influence of industrial enterprises and substantiation of approaches to the assessment of technogenic load on the environment;

– determination of the limits of applicability of the method for determination of the granulometric composition of fine coal materials by laser diffraction for purposes of environmental and process monitoring;

– investigation of environmentally significant characteristics (heavy hydrocarbons and sulphur compounds contents) of the gas extracted from coal-beds.

To address these objectives, a number of the most modern chemical and physicochemical research methods was used: inductively coupled plasma atomic emission spectrometry, atomic emission spectrometry in a variant of sample evaporation from electrode, atomic absorption spectrometry with electrothermal atomization, cold vapour atomic absorption spectrometry, GC-MS, HPLC, GC/GLC, laser diffraction, infrared spectrometry, scanning electron microscopy, X-ray phase analysis, low-temperature nitrogen adsorption method, energy dispersive X-ray spectroscopy, potentiometry, and photocolorimetry. The indicated methods were implemented using both standardized techniques and techniques developed in the measurements.

Toxic nitro compounds

Toxic nitro compounds including 2,4,6-trinitrotoluene are among the priority organic pollutants to be controlled in the area of operation of coal mining and coal processing industry enterprises.

Breaking rocks by means of the explosive energy is the universal and almost sole highly effective method of preparation of rocks for excavation. Large charges of explosives are increasingly used in the mining industry when charges of 1000 t are simultaneously detonated in an open-work and those of 100 t in underground ones. Kuzbass annually uses 600 000 t of explosives and in practice, up to 2–3 % of this amount is scattered in the area of mining. As a consequence, coal production enterprises are one of the main sources of the emission of nitro compounds into the environment. The most important brisant explosive is TNT. It is notable for distinct toxic properties. Furthermore, a maximum permissible concentration (MPC) of 0.01 mg/dm^3 referring to Hazard Class II (highly hazardous) was established for this compound in water bodies for household and drinking purposes.

Twenty-five samples of surface and waste water, and also 12 samples of wastes were analysed during 2006–2010. Table 1 gives the distribution of TNT and its metabolites [2-amino-4,6-dinitrotoluene [(2ADNT) and 2,4-dinitrotoluene (DNT)] in waste water of a surface mine. It is demonstrated that waste quarry water contains high concentrations of TNT. Moreover, applied methods of sewage treatment do not always lead to a reduction of TNT content. There are also 2ADNT and DNT identified in waste water. They are more toxic than TNT [1].

Polyaromatic hydrocarbons

Polyaromatic hydrocarbons (PAH) refer to resistant ecotoxicants and coal organic mass components. The content of PAH in coal may reach up to hundreds, and in some cases to thousands mg per kg. The emission of PAH into the environment is probable during production, storage, and transportation, and also during high-temperature coal conversion. A PAH representative with a strong carcinogenic effect is benzo(a)pyrene. The most common and resistant hydrocarbons of this series

TABLE 1

Distribution of TNT and its metabolites in waste water of open-pit coal mining enterprise at the Olzherasskoye coal deposit

Place of taking water samples	TNT content, mg/dm^3	Identified nitrogen compounds
Area 1:		
Quarry water before filtering array	0.0091	n/d
Same before filtering array	0.0124	n/d
Area 2:		
Quarry water before filtering array	0.0690	2ADNT, DNT
Same before filtering array	<0.0016	n/d

Note. 1. TNT is trinitrotoluene; ADNT is 2-amino-4,6-dinitrotoluene; and DNT is 2,4-dinitrotoluene. 2. n/d means not defined.

are phenanthrene, fluoranthene, pyrene, and chrysene. These hydrocarbons are the predominant components of emissions of systems related to pyrolysis of organic matter and act as convenient tracers to assess pollution of the atmosphere, surface water, and soils [2]. The latter function as a specific accumulator, whereat PAH falls into resulting from the global transfer of emissions from anthropogenic sources and natural atmospheric inputs. Therefore soils are the most representative targets to explore accumulation of PAH, the availability of which may play an indicator role reflecting the presence of pollution source [3].

Kuznetsk Coal Basin has large reserves of coals of all grades, from long-flame to lean and samples of various grade coals (D, DG, GZHO, W, K, CS, OS) that are in total almost a complete series of metamorphic transformation were selected for exploration. The high-performance liquid chromatography method was used to determine the PAH content in the coals (Table 2).

The maximum total value for PAH was found for coals of K and KS grades (363.4–432.0 μg/kg). In the coal samples, benzo(a)pyrene was detected. Its content was found in the 3.6–47.6 μg/kg range. The maximum concentrations were determined for phenanthrene. Its fraction was 50 % of the total amount of identified PAH. Thus, phenanthrene is a priority PAH for various grade coals and can be used to assess the impact of coal mining and processing onto the environment [4, 5].

In order to comparatively compare the pollution level of the territory of polyaromatic hydrocarbons, it is required to assess their background content. Thus, when examining 23 soil samples in the territory of Tashtagol district, it was determined that all of them contained

TABLE 2

Content of polycyclic aromatic hydrocarbons (PAH) in samples of various metamorphic grade coals from Kuznetsk Coal Basin

Compound	Content, μg/kg								
	Coal brand								
	D	DG	GZHO	GZHO	ZH	K	CS	CS	CS
Phenanthrene	80.8	14.3	86.2	50.6	33.0	204.2	199.6	103.2	124.9
Pyrene	36.9	1.2	14.0	12.6	19.6	69.2	66.2	6.7	32.1
Benz(a)anthracene	32.0	9.7	21.6	5.2	7.8	16.4	34.2	4.3	3.1
Chrysene	<3.0	<3.0	<3.0	17.9	11.9	62.6	59.6	32.4	28.0
Benz(b)fluoranthene	<6.0	6.2	13.0	26.0	18.8	11.0	24.8	7.6	5.6
Benzo(a)pyrene	<1.0	3.6	10.6	6.6	4.4	<1.0	47.6	24.3	21.4
Total PAH content	149.7	35.0	145.4	118.9	95.5	363.4	432.0	178.5	215.1

Note. D, DG, GZHO, ZH, K, CS, OS mean long-flame, long-flame gas, gas fat lean, fat, coking, coking sintering, lean caking coal, respectively.

<0.002 mg/kg of fluoranthene, just as much pyrene, and <0.001 mg/kg of chrysene. Phenanthrene was present in 10 samples in the concentration range from 0.001 to 0.005 mg/kg, with its background concentration in the soils of this territory (0.0019±0.0013) mg/kg. The paper [6] evaluated the distribution of PAH in samples of the soil of the Kemerovo region, systematized the results of the investigation of more than 300 soil samples. When examining the distribution of PAH in soils at the boundaries of sanitary protection zones of coal mining enterprises by the open and closed method, it is shown that phenanthrene content is in the range from 0.0012 to 0.0031 mg/kg. Herewith, benzo(a)pyrene is not detected in soils. A different distribution of PAHs therein is observed in the zone of influence of coal processing enterprises: phenanthrene content is found in the range from 0.0015 to 0.0432 mg/kg, the concentration of benzo(a) pyrene – from <0.001 to 0.0390 mg/kg (2 MPC). Thus, coal processing enterprises are sources of the ingress of PAH, including carcinogenic benzo(a)pyrene, into the environment.

However, processes for high-temperature coal conversion have the greatest impact on environmental pollution by compounds of the PAH class [7–10]. For example, Table 3 presents levels of soil pollution by phenanthrene in various functional zones of the central district of Novokuznetsk.

Phenanthrene content in liquid wastes from coke and by-product process is 66.0 g/kg. This type of wastes is a source of secondary environmental pollution by compounds of the PAH class. Phenanthrene content in the territory of the industrial site of the enterprise, the process cycle of which is associated with PAH emission (coke and by-product process of the Kuznetsk metallurgical combine), is 465 times higher than the background value.

Table 4 gives depth distribution of PAH and assessment of soil pollution category in the territory of the industrial site of the enterprise for high-temperature coal conversion.

Toxic elements and organic matter in solid wastes from coal production and conversion

Exploration of overburden and host rocks. Mining wastes cause serious environmental problems in the territory of the Kemerovo region. For example, 2661.281 million t of production and consumption wastes was formed at this territory in 2013. Among them, 2593.780 million t (97.46 %) falls on wastes formed during coal mining. Issues of exploring the composition of carbon waste to assess their toxicity and determine the list of priority elements to be environmentally monitored in the storage areas are important [11–13]. Research objects were overburden and host rocks (60 and 52 samples, respectively) formed during coal mining in the territory of the Kemerovo region. Reference [14] gives content ranges of gross forms of toxic elements in the overburden and host rocks.

Table 5 gives the ranges of the content of mobile forms of toxic elements in the overburden and host rocks. There is the maximum excess of MPC in mobile forms of elements in overburden rocks for Cu, Ni, Zn, and Pb (5.7, 3.4, 3.6, and 8.5, respectively). The maximum concentrations of mobile forms of heavy metals in host rocks are higher than MPC for Cu, Ni,

TABLE 3

Level assessment of soil pollution with phenanthrene in various functional areas of the central district of Novokuznetsk

Functional areas	Phenanthrene content, mg/kg	Multiplicity of background value excess, times
District recreational area. “Vodnaya” beach	0.008	4
Sanitary protection zone, Kuznetsk metallurgical combine enterprise	0.035	17.5
Area adjacent to highway Motor-road Oktyabrsky Ave. – Transportnaya Str.	0.155	77.5
Industrial site territory Kuznetsk metallurgical combine	0.930	465
Territory of testing ground liquid waste from coke production, Kuznetsk metallurgical combine	66 000.0	$33 \cdot 10^6$

TABLE 4

Depth distribution of PAH and assessment of soil pollution category in the territory of the industrial site of the enterprise for high-temperature coal conversion

Compound	PAH content, μg/kg				
	Sampling depth, m				
	0.0–0.2	0.2–0.5	0.5–1.0	1.0–2.0	2.0–3.0
Phenanthrene	336.22	100.16	23.63	66.72	11.15
Anthracene	78.92	20.03	3.42	4.66	2.68
Fluoranthene	1028.34	342.66	68.62	70.00	40.86
Pyrene	1037.42	345.80	67.32	62.40	42.77
Benz(a)anthracene	796.97	266.60	51.30	43.92	30.67
Chrysene	664.7	221.30	44.71	43.01	29.88
Benz(a)pyrene	540.97	180.45	36.62	38.31	26.25
Dibenzo(a,h)anthracene	190.63	50.22	<0.02	<0.02	<0.02
Benz(b)fluoranthene	428.24	142.26	29.50	33.74	22.10
Benz(k)fluoranthene	381.32	120.15	27.48	9.45	15.44
Benz(g,h,i)perylene	227.06	70.40	38.17	16.94	7.99
Indeno(1,2,3-cd)pyrene	299.63	98.50	45.02	23.82	14.03
Total	6010.42	1958.53	435.79	412.97	243.82
Soil pollution grade	Extremely hazardous	Extremely hazardous	Hazardous	Hazardous	Permissible

Zn, and Pb (by 7.9, 5.3, 2.0, and 3.8 times, correspondingly, Fig. 1).

The maximum concentrations of water-soluble forms of toxic elements in overburden rocks derived by water extraction are higher than MPC for water of fishery water bodies in 85, 80, 125, 21, 3, 50, 3, 20, and 5 times for V, Cu, Mo, As. Ni, Zn, Pb, Mn, and Cr, respectively. There is also a similar distribution of MPC by the same elements for the host rocks: 20, 200, 325, 3, 20, 23, and 3 for V, Cu, Mo, Ni, Zn, Mn, and Cr, respectively (Fig. 2). There are no Cd, Co, Sb in water-soluble forms of overburden rocks. The water-soluble form of mercury in overburden rocks is not detected, and in the host ones, its maximum concentration reaches 20 MPC. Thus, overburden and host rocks of Kuzbass coal mines are a source of toxic elements and are characterized by the activity of their water-soluble forms, whereat

TABLE 5

Content range of mobile forms of toxic elements (mg/kg) in overburden and host rocks of Kuzbass coal enterprises

Element	Content, mg/kg		MPC for soil, considering background, mg/kg
	Rocks		
	Overburden (n = 60)	Host (n = 52)	
Copper	3.0–17.2	3.0–23.8	3.0
Nickel	5.0–13.6	7.3–21.0	4.0
Zinc	19.0–82.4	6.1–49.9	23.0
Lead	10.0–51.7	2.0–22.6	6.0
Manganese	31.0–539.0	26.0–510.8	60–140
Chrome	<2.0–4.5	<2.0–5.4	6.0
Molybdenum	0.2–1.2	<0.05–1.8	–
Cobalt	0.08–0.1	0.08–0.12	5.0

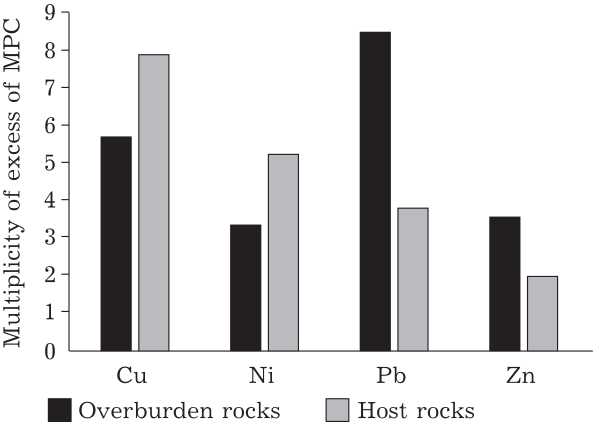

Fig. 1. Maximum permissible concentration (MPC) of mobile forms of toxic elements in overburden and host rocks.

the following series is determined: Mo > Cu > V > Zn > Mn > As > Cr > Ni > Pb.

The research findings allow concluding that pollution of water bodies of the Kemerovo region by such heavy metals, as V, Cu, Mo, Ni, As, Mn, Cr, and Zn takes place among other due to washing out during filtration of natural water flows through a large body of overburden and host rocks.

Issue of fine coal materials. Kuzbass heat and power industry enterprises have a great impact on environmental pollution of the region. The ash and slag waste (ASW) production is a severe issue of coal-based power generation. There was produced 2283.25 thousand t of ASW in 2014 and only 96.45 thousand t (4.22 %) there from was used. The total accumulated mass of BA is more than 88.8 million t. The problem becomes complicated by the limited scope of ash disposal areas and their expansion need, which results in alienated land resources and their pollution. The chemical composition data for ASW, toxic elements content and their ability to leach are the basis for the prediction of environmental pollution and the effective and environmentally friendly use of these wastes in the manufacture of construction materials, highway engineering, *etc.* [15–19].

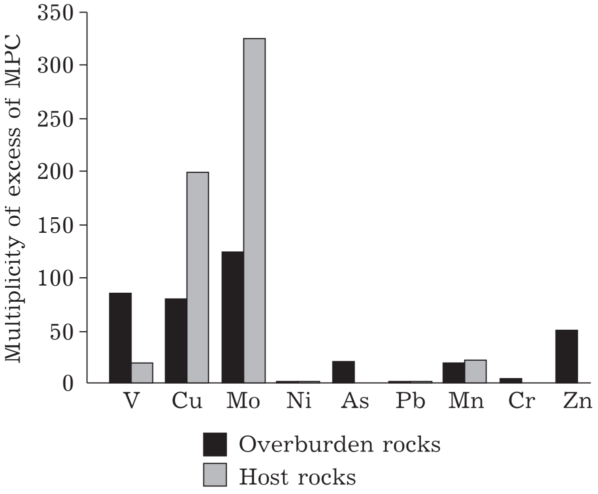

Fig. 2. Maximum permissible concentration (MPC) of water-soluble forms of toxic elements in overburden and host rocks.

Table 6 presents ranges of the gross content of toxic elements and compounds and oxides of ash-forming elements in BA from Kuzbass coal combustion. The systematization was performed according to the number of samples $n = 59$. As demonstrated by the research findings, among the toxic elements, the largest fraction falls for sulphur and manganese that are on the average 53.8 and 37.6 % of total toxic elements, respectively. The main oxides of the ash-forming elements are silicon dioxide (55.2 %), aluminium oxide (22.13 %) and iron trioxide (8.5 %). The residual carbon content varies widely and averages a significant amount (33.41 %). The average concentration of oil products in ASW is 167.8 mg/kg. Nitrates are found in ash and slags; their concentration varies in a wide range of concentrations, i.e. from 1.18 to 468.0 mg/kg.

In the course of exploring the interrelation between the content of individual elements in ash and slags, the reverse correlation between carbon and silicon dioxide, and also a correlative relationship between silicon dioxide and potassium oxide ($r = 0.72$) were found. The content of mobile forms of toxic elements was additionally determined. There are correlations between cobalt and nickel ($r = 0.85$), and also fluorine and zinc ($r = 0.59$) for mobile forms of elements. Furthermore, data analysis demonstrated that there was a correlation between the gross content and mobile forms of manganese and lead whereas being weak for copper and zinc (Fig. 3).

It is demonstrated that in contact with water, ash and slag waste is a source of toxic elements (vanadium, molybdenum, arsenic, nickel, zinc, manganese, and chromium) in groundwater in the territory of ash and slag dumps

TABLE 6

Gross content of toxic elements and compounds in bottom ash waste from Kuzbass coal combustion

Element, compound	Content, mg/kg	
	Range	Average
Elements		
Cadmium	0.05–0.40	0.32
Lead	4.07–310.0	94.89
Vanadium	7.13–179.27	57.35
Mercury	0.1–9.0	0.47
Arsenic	0.5–44.0	5.07
Chrome	5.50–150.68	55.36
Manganese	292.20–6585.15	1361.01
Antimony	0.17–5.82	1.55
Total sulfur	100.0–10 700.0	1675.9
Copper	6.55–66.27	20.87
Zinc	8.0–180.0	29.58
Nickel	7.67–44.61	25.92
Tin	1.00–6.00	3.25
Cobalt	2.83–13.54	7.44
Molybdenum	4.00–7.00	4.50
Compounds		
Benz(a)pyrene	0.001–0.1	0.0036
Mineral oils	20.0–5000.0	167.80
Nitrates	1.18–468.0	23.56
Formaldehyde	0.05–6.76	1.02
Phenols	0.01–0.05	0.011
Synthetic surfactants	0.20–10.47	0.87

and near natural water sources. The content of these elements in observation wells is significantly higher than their MPC in water.

Ash and slag from Kuzbass coal combustion should mainly be regarded as a source of the ingress of toxic elements (Mn, S, As, Cr, Cu, Zn, Ni, *etc.*) into the environment. Nitrite monitoring in environmental objects in the territory of storage of ASW is also relevant.

Issue of fine coal materials. The investigation of the granulometric composition of coal materials is of great importance in process, environmental and sanitary-hygienic monitoring. Utilization of coal industry wastes by the development of time-stable coal-water fuel requires monitoring the granulometric composition of coal sludges. An indispensable prerequisite during the modernization of emission purification systems of dressing plants, thermal generating units, and coal loading and unloading points is the information about the industrial dust particle size. The search for optimum engineering wastewater treatment solutions of coal enterprises from suspended matter, enhanced work efficiency of sludge pits also requires the information on particle granulometric composition and computation of precipitation curves. As coal enterprises are a source of suspended matter ingress into work area air and atmospheric one, the issues of particle size determination in environmental and sanitary and hygienic monitoring and inspection are of great importance. There is also a need for the inventory of industrial sources of atmospheric emissions of suspended particles with sizes less than 10 μm (PM10) and 2.5 μm (PM2.5).

The measurement of particle size distribution with the determination of the fraction of the most hazardous classes became possible with the appearance of the laser diffraction method being most informative and reliable.

Standard samples of GZhO, OS, and G coals (coal 1, 2, and 3, respectively) crushed to a size

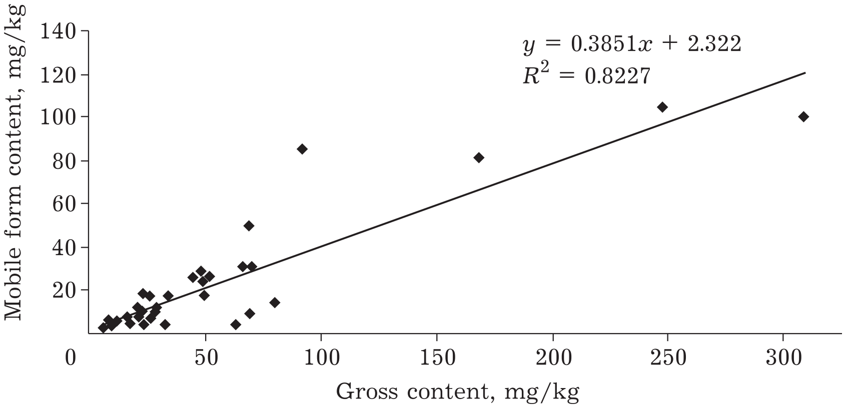

Fig. 3. Lead content at gross and mobile forms in bottom ash wastes for sample series.

of 0–0.2 mm were selected as research objects. Furthermore, the granulometric composition of coal sludge samples and dust emissions from dressing plants was determined [20, 21].

Granulometric composition homogeneity of standard samples of coal in sizes of –0.074...0 mm, –0.1...–0.074 mm, –0.2...0.1 mm was explored for 20 samples of each example. To determine the granulometric composition of coal powders, there were used two methods: sieve analysis according to GOST 2093–82 and laser diffraction. The FRITSCH ANALYSETTE 22 MicroTec Plus size analyser (Germany) was used for measurements by the laser diffraction method.

As demonstrated by the results acquired using this procedure, the emissions of dressing plants contain more than 50 % of particles, most dangerous to the respiratory system, with sizes in 10 μm (Fig. 4) and therefore treatment is required.

Gaseous pollutants entering the atmospheric air from coal-beds

Methane and carbon dioxide from coal-beds make a significant contribution to the composition of greenhouse gases [22]. During the period from 2011 to 2013, the determination of the content of these gases in the atmospheric air at the boundaries of sanitary protection zones (SPZ) of mobile degassing units (MDU) of coal mining enterprises was carried out. Altogether more than 500 samples of the atmospheric air were analysed in the borderline of SPZ of SDW landfill and in the atmospheric air of populated areas. Methane content ranges in the atmospheric air from 0.31 to 143.61 mg/m^3 (within the boundaries of SPZ of MDU) and between 1.08 and 3.89 mg/m^3 (in the borderline of SPZ of SDW landfill), and from 0.94 to 2.28 mg/m^3 (in the territory of settlements). Carbon dioxide concentrations are found within 0.043 and 0.065 % (in the borderlines of SPZ of MDU) and 0.038–0.049 % (in the borderline of SPZ of SDW landfill).

As demonstrated by the investigation of gas componential composition in 7 wells of the Taldinsky coal mine, methane content therein lies at a level of 91.95–98.26 %. The concentration of heavy methane hydrocarbons varies in a wide range of concentrations (ethane from 0.007 to 2.109 % and propane from <0.001 to 0.401 %), the content of butanes, pentanes, and hexanes in the gas is insignificant or not manifested [23–26].

Hydrogen sulphide concentration in coal-bed gas of the Naryk-Ostashkin area lies in the range from 0.00015 to 0.0054 mg/m^3.

DEVELOPMENT AND INTRODUCTION OF TECHNOLOGIES TO REDUCE ENVIRONMENTAL POLLUTION

Catalytic fluidised bed fuel combustion

Catalytic heat generators (CHG) were developed at the BIC SB RAS to provide environmentally friendly and efficient fuel combustion [27]. Fuel oxidation in CHG proceeds on the surface of spherical granules of special oxide catalysts maintained in a fluidised state in a flow of fuel, air, and combustion products.

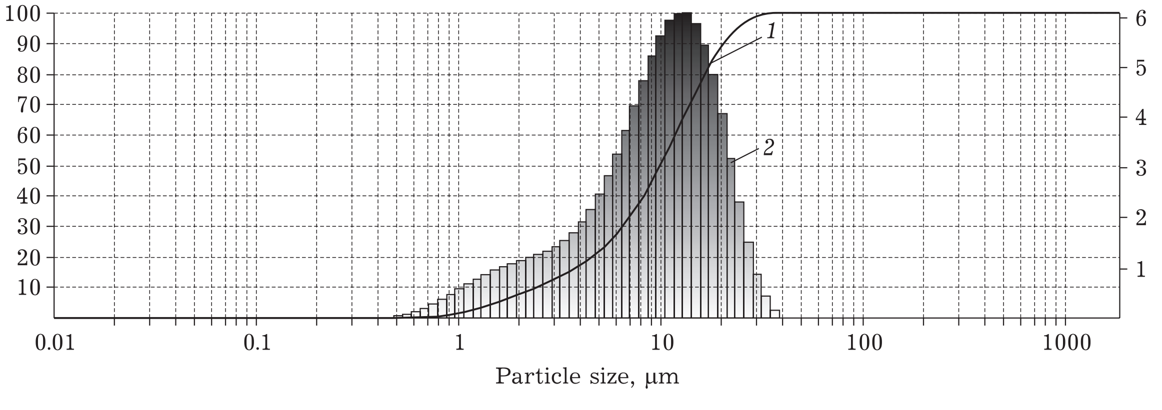

Fig. 4. Integral (*1*) and differential (*2*) particle size distribution curves of coal dust in industrial emissions into the atmosphere of concentrating mills. Distribution parameters: D_{10} of 2.4 μm, D_{50} of 9.9 μm, D_{90} of 20.3 μm, D_{99} of 30.0 μm. Notation: *Q* is volume ratio of particles, which size is less than the current one, d*Q* is a fraction of particles of a given size; D_{10} is particle size, less of which 10 % of particles is present, D_{50}, D_{90}, and D_{99} are sizes, less of which 50, 90, and 99 % of particles is found.

TABLE 7

Typical composition of coal-bed gas of Taldinsky coal mine for various wells (UM-5.4, UM-5.5, UM-5.6, UM-5.7, UM-5.8, UM-5.9, UM-5.10), vol. %

Component	Well number						
	UM-5.4	UM-5.5	UM-5.6	UM-5.7	UM-5.8	UM-5.9	UM-5.10
CO_2	0.199	0.922	0.464	0.736	0.455	1.182	1.836
O_2	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005	<0.005
N_2	0.861	0.232	0.728	0.395	0.333	0.866	0.846
CH_4	98.17	96.36	97.66	98.26	98.17	97.20	97.11
C_2H_6	0.007	0.861	0.006	0.017	0.009	0.008	0.046
C_3H_8	<0.001	0.157	<0.001	<0.001	<0.001	<0.001	0.011
i-C_4H_{10}	<0.001	0.011	<0.001	<0.001	<0.001	<0.001	0.002
n-C_4H_{10}	<0.001	0.002	<0.001	<0.001	<0.001	<0.001	0.002
i-C_5H_{12}	<0.001	0.002	<0.001	<0.001	<0.001	<0.001	<0.001
n-C_5H_{12}	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
C_6H_{14}	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001	<0.001
H_2	<0.001	0.001	<0.001	<0.001	<0.001	<0.001	0.001
He	0.003	0.008	0.004	0.006	0.006	0.002	0.001

Heat is removed from the catalyst bed through heat exchange surfaces located in the bed or by direct contact of the catalyst with the working medium [27–30].

As demonstrated by the research carried out for combustion of various fuels in CHG, the use of the latter allows eliminating the formation of products of incomplete combustion, such as carbon black and carcinogenic hydrocarbons, and significant reduction of CO and NO_x emissions. Comparison of NO_x concentrations during flame and fluidised bed combustion demonstrated that fuel combustion in CHG resulted in a drastic reduction of the formation of both thermal and fuel NO_x [28–30].

Fundamental research on catalysts synthesis and investigation of reactions of complete catalytic oxidation allowed recommending CHG devices to perform various technological processes. The latter are currently being introduced into the industry.

The use of CHG is most efficient in the following processes [31–33]:

– heating and evaporation of liquids and heat supply;

– drying of powder materials including coal;

– adsorption-contact drying of temperature-sensitive products of chemical manufacture with adsorbent regeneration in CHG;

– catalytic treatment of highly concentrated gas emissions and organic waste with heat recovery;

– catalytic treatment of dust-loaded gas emissions from coke dry-quenching plants that contain CO and hydrogen;

– combustion of low-calorific fossil fuels that are unsuitable for flame burning;

– conversion of brown coal from the Kansk-Achinsk Basin with simultaneous production of semi-coke and synthesis gas.

Efficient and environmentally friendly boiler stations operating on liquid [34, 35] and solid fuel [35, 36] were developed on the basis of CHG.

Original developments in the area of catalytic fluidised bed combustion are aimed at the destruction of hazardous organic waste, such as the highly toxic liquid rocket fuel 1,1-dimethylhydrazine [37] and mixed organic waste containing radionuclides [12, 13]. The process for catalytic oxidation of 1,1-dimethylhydrazine in a pilot installation was developed with participation of Academician V. P. Makeyev State Rocket Centre (Miass city). The BIC SB RAS together with a number of Minatom enterprises of the Russian Federation elaborated a process and a pilot plant for catalytic treatment of mixed organic waste with heat recovery. The plant was manufactured at the Novosibirsk chemical concentrates plant, PJSC [38, 39].

The CHG technology based on the use of a fluidised bed demonstrated high efficiency in gas purification from hydrogen sulphide by its direct oxidation to elemental sulphur [40–44]. Obvious advantages of this technology are the continuity of the process, which allows simultaneously combining gas purification with the production of commercial sulphur; soft conditions of the process through the use of specially developed high-active catalysts (T = 220–280 °C); simplicity, and flexibility of the process control. An opportunity to effectively remove heat from the catalyst bed allows removing restrictions on maximum hydrogen sulphide concentration in the processed gases and effective utilization of the reaction heat.

Honeycomb catalysts

Monolithic catalysts of the honeycomb structure (cellular structure with parallel channels of certain geometry with thin dividing walls between them) have several unique features: a substantial uniformity of the geometric structure, high ratio of surface to volume, low pressure drop and high mechanical strength and thermal stability. It is most justified to use honeycomb catalysts in processes with high exothermic effects at low contact times and high space velocities. Honeycomb catalysts are widely used in the practice of gas purification in Western countries. The well-known companies Corning, Engelhard (USA), Siemens, Degussa, BASF (Germany), NGK (Japan), Haldor Topsoe (Denmark), and others produce monolithic cellular carriers and catalysts [45].

Paper [46] lists the main areas of promising research trends of the BIC SB RAS on the development of monolithic honeycomb catalysts for environment protection processes. In accordance with the above areas, efficient catalysts were developed for:

– direct single-step oxidation of hydrogen sulphide to elemental sulphur [47] and the decomposition of hydrogen sulphide including that in the presence of ammonia [48–50];

– selective catalytic reduction of nitrogen oxides by ammonia [38, 51];

– same by hydrocarbons [52–54];

– oxidative coupling (dimerisation) of methane [55];

– environmentally friendly catalytic fuel combustion [56–60];

– purification of diesel engines exhaust gases [61–64];

– treatment of waste gases of enterprises from hydrocarbons and CO by catalytic afterburning [59, 65–67].

Reference [68] analyses and compares toxic impurities contained in a raw coke gas in foreign and Russian by-product coke industry enterprises. The above-described processes of purification of coke gas are divided and classified from the standpoint of physicochemical fundamentals and resulting products. It is demonstrated that the coke-chemical industry requires the development of modern sulphur recovery units. The process for the direct oxidation of hydrogen sulphide over honeycomb catalysts developed at the BIC SB RAS is quite an effective method to address this issue.

A crucial environmental issue is the combustion of waste gases including those of high pressure during recovering and refining natural gas and oil. Flaring facilities, jumbo burners, and other flare combustion devices are used for that purpose. During flaring, the amount of hazardous compounds is as follows: carbon monoxide, incomplete oxidation products (benz(α)pyren and aldehydes), nitrogen oxides, and carbon black is 25–50 kg per ton of emissions. The quantity of emissions increases to 100 kg per ton of fuel upon burst releases performed during the planned or emergency shutdown of reactors. To disperse harmful combustion gases in the atmosphere both routinely and especially during burst releases, there is a need to create high (up to 100 m) flare constructions, due to which maintenance and online repair of flare devices are difficult.

This problem may be solved efficiently by creating an installation containing two different diameter catalytic elements based on monolithic honeycomb catalysts with an opportunity to involve one or two of them depending on gas yield [69].

An original area of the use of honeycomb catalysts is the technology of two-step combustion of hydrocarbon fuel. In the first step, the fuel is burned in the flame burner under conditions of a small lack of oxygen to suppress the formation of toxic nitrogen oxides. In the second step, the combustion products are diluted with air, and the hot gas is fed to the catalytic cassette from the ceramic monoliths of the cellular structure, where products of incomplete burning are oxidised and micro-impurities of nitrogen oxides are abated to sanitary standards. Along with environmental purity, these catalytic heaters have an important advantage, such as fuel KPI (Key Performance Indicator) close to 100 %. The use of such devices is most efficient for heating glasshouses, as along with the heat-up, there is a simultaneous feeding of plants with carbon dioxide [59].

Catalytic technologies for abatement of nitrogen oxides in flue gases

Heat power engineering is one of the main sources of atmospheric technogenic pollution by harmful substances: nitrogen oxides (NO_x) and sulphur oxides (SO_x), and also solid particles [28, 29, 70]. Nitrogen oxides make a great contribution to the formation of photochemical smog, acid rain, ozone layer destruction, and the greenhouse effect, therefore a decrease in their emissions is quite an urgent task.

From the technological and economic standpoint of gas treatment processes, the most efficient method for pollutant abatement to the MPC level is catalytic processes [71–73]. The selective catalytic reduction (SCR) process of nitrogen oxides is one of the most promising methods to prevent environmental pollution. The main reaction products are harmless nitrogen and water vapours. The SCR processes are carried out in the presence of reductants, such as CO, H_2, ammonia, or hydrocarbons. The most commonly used process for reduction of nitrogen oxides is SCR with ammonia. Advantages of this technique include a relatively low cost and a high efficiency. The NOx transformation degree may reach 95 %. This method is used to purify gases from stationary sources, such as energy cycle enterprises.

The most known catalyst currently used in NH3-SCR industrial processes is WO_3 or MoO_3 promoted V_2O_5 supported on TiO_2 [74, 75]. The operating temperature of catalysts is 300–400 °C. In practice, for abatement of nitrogen oxides in flue gases, monolithic catalysts with honeycomb structure are used. The catalysts used in the NH_3-SCR process have unique functional characteristics, such as the high mechanical strength, the low pressure drop, and resistance to the precipitation of carbon, dust, and metals during NO_x removal process, and usability. A homogeneous mixture of titanium oxide, tungsten oxide or molybdenum oxide and vanadium (V) oxide, which is moulded into monoliths of honeycomb structure, is usually used as a commercial SCR catalyst for thermal stations [76]. The monoliths are assembled into standard modules that are placed inside the reactor to form catalytic layers.

Reference [51] considers methods to prepare honeycomb catalysts containing the active component V–Ti–O, Cu–Ti–O, Cu–ZSM-5, and Co–ZSM-5 on the washcoat layer on the surface of monolithic ceramic and oxide carriers of the honeycomb structure. It is demonstrated that activities of the examined samples under certain conditions of active component formation are comparable to those of bulk monolithic catalysts produced by extrusion.

As demonstrated by the tests of the honeycomb oxide catalysts IK-44, IK-45, IK-46, IK-47 developed at the BIC SB RAS, during purification of exhaust gases from the plasma incinerator of waste (Butte, Montana, USA), catalysts of the BIC SB RAS similar by the activity to the western commercial SCR catalyst ensuring the degree of purification at a level of 90–95 % [39]. In addition to that, these catalysts have an advantage in the environmental safety of their utilization. The leaching test of metals in their composition demonstrated that the level of the leached metals was significantly lower compared to testing results of commercial catalysts.

The use of hydrocarbons as reducing agents instead of ammonia has the following obvious advantages:

– there is no need to build a hazardous reagent (ammonia) warehouse;

– catalytic reactor design is simplified due to the reduced requirements for unreacted ammonia slip;

– a less expensive reducing agent is used.

Metal-substituted zeolites, in particular, CuZSM-5 are used as catalysts for the SCR process by hydrocarbons. The optimum content and the most active cationic states were studied in detail on the basis of bulk catalysts by systemic variation of catalyst preparation conditions in a broad value range at the BIC SB RAS [76–81]. The purification efficiency in the presence of honeycomb Cu–ZSM-5-containing catalysts may reach 70–80 % at a temperature of 300–600 °C and a space velocity of 30 000–45 000 h^{-1} [52–54]. A disadvantage of zeolite catalysts is their deactivation in the presence of water vapour and sulphur oxides. The supported Cu–ZSM-5 catalysts modified with cerium oxide and titanium oxide developed in the BIC SB RAS show high resistance towards poisoning by sulphur compounds and water vapour and may be recommended for practical use for real flue gas treatment [82, 83]. Furthermore, it was shown that Cu–ZSM-5 catalysts were active in carbon black oxidation in a NO/O_2 mixture [83].

Development of the scientific and technological base for the chemical processing of coal-bed methane

Coal production processes result in the released coal gas that is contained in a coal-bed and surrounding rocks. Kuzbass mines annually emit ~1 billion m^3 of methane [84]. However, only a minor part (below 3 %) of coal-bed methane (CBM) is used for local gas feeding. Atmospheric methane emissions result in the enhanced greenhouse effect and the destructed Earth's ozone layer. As a greenhouse gas, methane has a high degree of impact on climate change: the greenhouse effect of methane is ~25 times higher compared to carbon dioxide. Apart from environmental pollution impact, methane release into mine workings is one of the limiting factors of enhanced coal production.

The total coal-bed methane resources in the main coal-bearing basins are assessed as 83.7 trillion m^3. Whereas Kuznetsk Coal Basin (methane reserves of 13 trillion m^3) is referred to highly promising coal fields that maximally meet the requirements for large-scale production of CBM [85]. According to quality indicators, coal-bed gas is insignificantly different from traditional natural gas and is characterised by methane content in 97–99 %. The CBM may be used in two main trends, such as energy (heat and electricity production) and chemical (synthesis of basic products and intermediates of the chemical industry). The CBM production at an industrial scale in Russia and technologies for its further processing lie at the initial step of their development.

With the aim of creating the scientific and technological base of the chemical processing of CBM into highly valuable commercial products, special catalysts were developed to prepare the following compounds from coalmine methane:

– benzene and hydrogen by methane dehydroaromatization [87–97];

– ethylene by oxidative coupling (dimerisation) of methane [98–102];

– hydrogen-containing gas (synthesis gas) by autothermal [103–116] and carbon dioxide [117–123] methane reforming.

During the development of highly active and stable catalysts for conversion of coal mine methane into highly liquid products of the chemical industry, a systematic research of regularities of material formation and impact of various synthesis parameters and reaction conditions on target products yield was carried out [87–116]. The preparation method, carrier composition, active component formulas, modifying additives type and content, thermal treatment and activation mode, and also conditions of catalytic processes were optimised.

Methane dehydroaromatization over metal-zeolite catalysts opens up an opportunity for direct production of expensive value-added products, *i.e.* benzene and hydrogen, from CBM [87–97]. Non-oxidizing conditions of the process ensure a high selectivity of target products formation (no less than 70 %). Furthermore, methane conversion is accompanied by the formation of hydrogen that is a valuable raw material for hydrogen energetics and the main reactant of petrochemical manufacturers (hydrotreatment of oil fractions and hydroreforming). The experimental sample of the composition 2Mo0.5Co/80ZSM-5/20CaM moulded in granules is recommended as the optimum catalyst for a one-step process of catalytic methane conversion to benzene and hydrogen [97].

The oxidative coupling of methane (OCM) is the direct process of conversion of methane to C_2-hydrocarbons, *i.e.* ethane and ethylene. The latter is a basic semi-product of the chemical industry. It is widely used for the production of polymer materials and chemicals, such as acetaldehyde and synthetic ethyl alcohol. Under conditions of fast growth of ethylene production capacities, there is a need for the expansion of the raw material base by the attraction of the main component of coal gas – methane. Resulting from a systematic investigation of the effect of various parameters on OCM process characteristics, $2Mn1.6Na3.1W-2La/SiO_2$ catalyst that ensures a yield of C_2-hydrocarbons of 22 % in methane conversion of 54 % was developed [98–102].

Autothermal reforming (ATR) of methane is an efficient process to produce hydrogen-containing gas from CBM. Proceeding from the investigation of regularities of catalysts formation and their evolution during catalytic processes, a stable catalyst for the ATR process of CH_4 was developed. The catalyst $10Ni0.005Pd/10Ce_{0.5}Zr_{0.5}O_2/Al_2O_3$ is characterised by stable operation under conditions of the ATR reaction of CH4 ensuring an H_2 yield of ~70 % in CH_4 conversion of ~100 % [103–116].

Carbon dioxide reforming of methane may be regarded as an environmentally friendly reaction, as greenhouse gases, such as carbon dioxide and methane, are used as the main raw materials to produce hydrogen-containing gas [117–123].

A promising method for methane utilisation is its decomposition over solid catalysts with the production of environmentally friendly hydrogen fuel and products with a high added value, which are carbon nanofibers and nanotubes. The latter are widely used in the production of materials and composites with unique properties, fuel cell electrodes, and supercapacitors, in various areas of electronics, medicine, chemistry, and catalysis [124–143].

Thus, the scientific and technological base for the processing of coal mine methane to benzene, ethylene, and hydrogen-containing gas with the use of nanoscale catalysts based on oxide supports was developed with the aim of solving the problems of deep processing of natural resources and environmental safety.

Nature of coal-bed methane hydrate

The presence of CBM presents a great number of environmental issues, on the one hand, as a source of greenhouse gas, and on the other one, as a direct hazard to human life. To improve procedures for prediction and prevention of hazardous gas-dynamic phenomena in coal mines, and also environmentally friendly technologies for CBM and its further rational utilization, it is required to explore the nature of the bond of methane with coal solid matter [144–148]. Methane hydrate presents a form of CBM and is formed in the presence of water at a pressure of the hydrate-forming gas of 2–7 MPa or above, and also at a temperature not over 10 °C. When gas pressure decreases below the equilibrium curve, gas hydrates decompose, which is accompanied by abnormally high gas release and thermal effect.

Thermobaric points of gas hydrate decomposition in natural coal were experimentally measured in [144–146]. All points are close to the equilibrium curve of bulk hydrate, on the basis of which it is possible to make a conclusion regarding the lack of the effect of decomposition points displacement occurring in other porous media [147]. The amount of water and gas involved in the formation of the hydrate was measured for coal samples in the metamorphic series [144, 145, 148]. It was determined that there was a moisture threshold for each sample of natural coal, in excess of which methane hydrate was formed. The measured properties of hydrates formed in natural coal were compared with the probable impact of the decomposition of gas hydrates on the occurrence of hazardous gas-dynamic phenomena in coal-beds.

Carbon sorbents to address environmental issues of Kuzbass

The most unfavourable environmental conditions develop in the Kemerovo region, as and in the great majority of large industrial regions, where a large number of industrial enterprises are concentrated. Surface water in the region is experiencing great anthropogenic pressure being exposed to pollution with the sewage of the mining, fuel and energy, metallurgical, coking, chemical industry, woodworking industry, agriculture, and public service enterprises. The discharge of contaminated industrial water in the territory of the Kemerovo region is annually 680–760 million m^3. Herewith, 400–450 million m^3 of them are discharged into water bodies without treatment.

Effluents of chemical, coke, chemical, and pharmaceutical industries are of special hazard, as they mainly contain dissolved substances. According to FGUZ Federal Centre of hygiene and epidemiology in the Kemerovo region, I and II hazard class substances, including phenols, organochlorine compounds, formaldehyde, and a number of others, are found both in the Tom river and drinking water [149–151].

Another essential issue is the quality of the drinking water of the centralized drinking water supply system. The consumption of water that does not meet hygienic quality standards determines the poor sanitary and epidemiological condition of many rural settlements in

Russia. In total, 29.5 million people in the rural area use water that does not meet the quality standards, of which 9.0 million people receive non-potable water from decentralized sources and 20.5 million men – from centralized water supply systems.

The fraction of water samples from drinking water supply sources not meeting the hygienic regulations according to sanitary-chemical indicators at large, in the country is 28.4 % (including surface – 27 %); microbiological indicators – 9.1 (21.6 %).

To overcome the above problems and supply pure water to the Russian population, mainstreaming effective water purification systems and equipping them with modern carbon sorbents are required.

Currently, the world production of sorbents is about 1.9 million t (or $ 5 billion in monetary terms) per year with the prospect of growth by 2024 to a level of 5.1 million t per year. Sorbent consumption in Russia is around 20–25 thousand t per year, in other words, with the unsteady economic environment, on the order of 70 % of consumed sorbents is currently imported. The specific production of sorbents in Russia is currently 20 g/person per year, which is 25 times below the world level. Considering the latter, it is required to produce no less than 70 thousand t of active coal per year in Russia to steadily develop the economy and set up high-quality living standards.

The Institute of Coal Chemistry and Material Science of the Federal Research Centre of Coal and Coal Chemistry SB RAS carried out a series of research aimed at enhanced thermochemical transformation of coal using ozonation, mechanochemical and chemical activation methods. A number of chemical and thermocatalytic techniques were developed for modification and preparation of highly porous nanostructured carbon materials out of a wide range of carbon-containing raw materials.

The technology for preparation of highly porous carbon sorbents based on naturally oxidized coals by their carbonization in the presence of alkali has been developed. Coals at different stages of coalification may act as the initial raw materials to produce carbon sorbents with a developed specific surface area. Physicochemical properties of the porous structure (texture) of carbon sorbents have been explored integrally and their dependence on the nature of the precursor coal has been determined [152, 153].

It is demonstrated [154, 155] that the mechanoactivation effect on coal-alkali mixture followed by carbonization makes it possible to produce carbon sorbents with high values of the specific surface area (S_{BET} is 1400 m^2/g) and total pore volume ~1.1 cm^3/g. The process of sorption of phenol and other harmful organic compounds by carbon sorbents from aqueous solutions has been explored. Moreover, it has been determined that the resulting sorbents are efficient to treat drinking and industrial waste water from organic pollutants.

Currently, the Institute of Coal Chemistry and Material Science of the Federal Research Centre of Coal and Coal Chemistry, SB RAS is implementing the project “Development of technology of obtaining of effective sorbents, creation of the experimental-industrial stands of sorbent production in Russia and the Republic of Kazakhstan for water treatment and enhanced life quality” realized within the agreement with the Ministry of Education and Science of the Russian Federation (RFMEFI61317X0079) by the Federal Target Program “Research and development on priority areas of the development of the scientific-technological complex of Russia for 2014–2020”. The purpose of the project is to develop the scientific bases of technologies for the preparation of effective sorbents, directly from Kuzbass fossil coal to address environmental issues of the region itself in water treatment from various pollution including heavy metals, organic compounds, and oil products.

CONCLUSION

Resulting from intensive coal production and preparation, developed iron industry, produced ferroalloys and aluminium, and also the presence of large-tonnage chemical industries and coal-fired power industry, extremely unfavourable environmental conditions have developed in Kuzbass. This review gives the results of some research on the current state of pollution of air, water and soil monitored in Kuzbass. Of course, carcinogenic polyaromatic hydrocarbons, coal dust, by-product coke and metallurgy industry products, and also absolutely new pollution sources, such as 2,4,6-trinitrotoluene and its metabolites typical for this region, are of special concern. To reduce and prevent pollutants emissions and to treat the polluted environment, a series of technologies are suggested. They are developed at the Boreskov Institute of Catalysis SB RAS and the Institute of Coal Chemistry and Material Science of the Federal Research Centre of Coal and Coal Chemistry SB RAS.

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