Main Scientific Aspects of Black Carbon Emissions

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Abstract

The definition of the concept " carbon black" in accordance with modern world ideas was given. The structure of carbon black, the morphology of its aggregates and elemental composition were described in detail. The place of carbon black among other black emissions in the atmosphere was characterized according to international standards; the fundamental differences between carbon black and soot were shown. The sources of carbon black emission into the atmosphere and soil were given. Industrial methods of carbon black production − thermal, furnace and channel − were reported. Basic diagrams of setups for the recovery of carbon black from aerosol streams in the production process providing a maximum possible efficiency of effluent gas and wastewater purification were described. Schemes and principles of operation of the main elements of carbon black recovery systems − cyclones and filters − were presented. The detrimental effects exerted by carbon black on the environment as a whole and on the human body in particular were demonstrated and the maximum permissible concentrations of carbon black were listed. The behaviour of carbon black in the atmosphere and oil contaminated soils was described. Methods of carbon black conversion in the atmosphere and schemes of its oxidation were outlined. A series of model experiments was performed to reveal the effect of carbon black on oil contaminated soils. The beneficial effect of carbon black on the removal of oil contaminants from soils was established. It was shown that carbon black is able to catalyze the destruction of various organic compounds, particularly oil hydrocarbons, thus improving soil fertility. Aerobic deterioration of carbon black in soils with time was discussed.

Key words: carbon black, emission, oxidation of carbon black, oil contamination of soils, degradation of oil contamination

INTRODUCTION

Environmental protection and nature management are now among the most topical problems nowadays. Particular attention is paid to the harmful emission to the atmosphere caused by intensive industrial development, since the amounts of such emission significantly exceed the neutralizing ability of nature.

Among various emissions of chemical substances, carbon nanoparticles emissions are of special interest. They are retained in the atmosphere from several days to several weeks and belong to unstable compounds that promote climate changes. Such particles absorb solar radiation, which results in warming of the atmosphere and shifting of the rainfall schedule. Their deposition on the earth makes its surface more dark, thus decreasing its reflectivity and facilitating an intensive absorption of sunbeams. Within the International Convention on Long-Range Transboundary Air Pollution, black carbon was introduced into the list of pollutants [1].

Black carbon has a low thermal conductivity coefficient, $0.365-0.425$ W/(m · K) [2], and a small particle size, $10-500$ nm; this produces Rayleigh light scattering on its particles, so its emission to the atmosphere certainly threatens the thermal climate of our planet. Black carbon is especially dangerous for the Arctic zone because it changes the reflectivity of snow, thus facilitating it thawing and thinning of a snow cover [1, 3]. Thus, great emphasis on black carbon was made in the report of the Task

Force established according to the Declaration of ministerial session of the Arctic Council, which was held in Tromsø in April 2009 in order to assess the existing and emerging mitigation options for fugitive compounds leading to climate changes and to develop appropriate recommendations. On 24–25 April 2015, foreign ministers of the Arctic Council countries adopted the Iqaluit Declaration, which again stressed the necessity to reduce emission of black carbon [3]. This testifies to the importance of research in a relatively unexplored field of carbon particles emission and the necessity of technical studies aimed at the development of recommendations on their emission reduction.

It is known that black carbon can aggravate some respiratory and cardiovascular diseases. However, the toxicological assessment of the properties of carbon employed in cosmetics industry, which was made in the report of European Scientific Committee on Consumer Safety (SCCS) [4] and in [5] is more optimistic.

The family of carbon materials includes more than a dozen of carbon modifications; among them, carbon black, which is the product of incomplete thermal or thermal oxidative decomposition of hydrocarbons, is produced in greatest quantities [6]. Each year, more than 11 mil-

lion tons of carbon black is produced in the world at more than 100 plants, and its production rate is constantly increasing [7].

There are more than 100 grades of commercially available carbon black differed by physicochemical and morphological properties, which, in their turn, depend on the method of carbon black production, its feedstock and process conditions [2].

Huge capacities of carbon black production are related mostly to its unique reinforcement properties. More than 50 % of the produced carbon black is used to manufacture high-strength and wear-resistant tyres. The introduction of carbon black gives a tenfold or even greater increase in the strength of vulcanizates [8]. In addition, carbon black is intensively consumed as a pigment in the printing industry as well as in the production of various composites and special-purpose mechanical rubber goods. At present, there is no alternative to carbon black, so its annual production is growing.

The manufacture of carbon black is inevitably accompanied by its emission to the environment: to the atmosphere with industrial off-gases and to water reservoirs with wastewater. Carbon black is distributed in the environment and under certain conditions can adversely af-
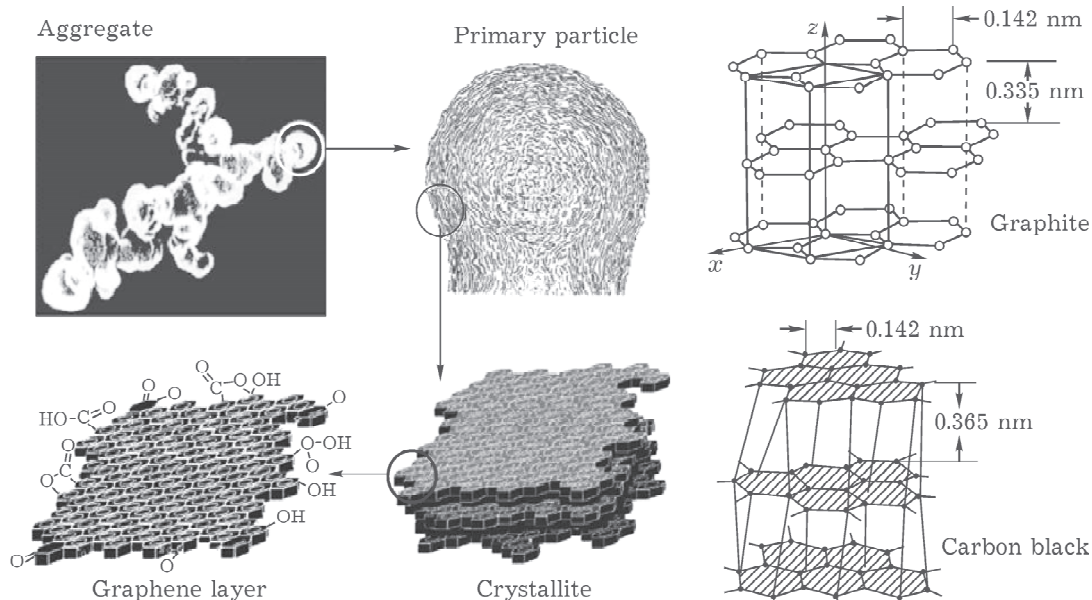

Fig. 1. Structural scheme of carbon black.

fect the living organisms and the entire ecosystem [9]; in this connection, careful control of industrial wastes from the carbon black production is needed.

PLACE OF CARBON BLACK AMONG OTHER BLACK EMISSIONS

The primary and indivisible particle of carbon black is its aggregate − a clustered entity consisting of 10−700 primary particles carbonized at the sites of coagulation contacts [6]. The primary particles are represented by a set of crystallites comprising 3−5 graphene layers with various functional groups that may reside on their terminal atoms [8] (Fig. 1).

In distinction to graphite, carbon black has two-dimensional ordering instead of three-dimensional one; its graphene planes in the crystallite, although being approximately parallel to each other, are shifted with respect to adjacent layers [8]. According to current concepts, graphene planes in carbon black primary particles of different size are bent, deformed and intercross to connect the adjacent primary particles. The lowest degree of ordering of graphene planes in the carbon black particle is observed in its nucleus and increases toward the surface. Such structural type is called paracrystal [8].

In some publications, carbon black is referred to as soot; however, these are absolutely different notions. In compliance with the ASTM D6602 standard, a reasonably selected set of methods, including transmission electron microscopy (TEM), TGA analysis and X-ray diffraction analysis (XRD), was proposed to differentiate carbon black not only from soot but also from other black emissions present in the environment. The main information is provided by TEM, which shows the detailed morphology of carbon materials at a high magnification. According to the mentioned standard, soot and carbon black, unlike other black emissions, are carbon materials with the clustered morphology; however, a detailed examination reveals strong differences between soot and carbon black [10].

First, carbon black differs from soot in its microstructure. It has a turbostratic structure, *i. e.* the external graphene layers are oriented concentrically with internal ones, whereas in soot the graphene layers are oriented in paral-

lel, without concentric orientation [10−12]. Primary particles of carbon black have virtually a smooth perimeter, while the perimeter of soot particles is much more crooked and irregular. Carbon black primary particles have a narrow size range within each grade (10−100 nm for furnace grades and 200−500 nm for thermal ones), whereas in soot the size of primary particles can widely vary even for the same sample. In addition, in carbon black the necks connecting primary particles are always smaller than the particles, while in soot they have comparable sizes [10].

Distinctions between soot and carbon black are observed also in their elemental composition. The fraction of carbon is much greater in carbon black in comparison with soot and commonly reaches 97 %; the only exception is the strongly oxidized channel carbon black in which carbon may constitute only 93 % [2]. Sulphur is absent in thermal carbon black grades but present in the furnace ones; depending on the feedstock, its weight fraction does not exceed 2 %. For carbon black, the concentration of volatile substances released during its calcination at 950 °C for 7 min is commonly not higher than 8 %. In the case of soot, the amount of sulphur varies in a wide range and the concentration of volatile substances is not lower than 20 % [10].

In addition, the H/C ratio in carbon black is closer to that in graphite in comparison with soot. For example, in electrically conducting carbon black the H/C ratio does not exceed 0.02, while in diesel soot it is near or above 0.3 [2].

SOURCES OF CARBON BLACK

The main sources of carbon black emission to the environment are carbon black enterprises. Several industrial methods are employed for the production of carbon black and each of them yields its modifications with specific properties and makes different contributions to carbon black emission to the environment [8, 13].

The thermal method (Fig. 2, *a*) is based on the decomposition of the gaseous raw materials without the access of air to the channels of refractory packing of a generator. The feedstock is delivered to the bottom of generators together with air and burned until the temper-
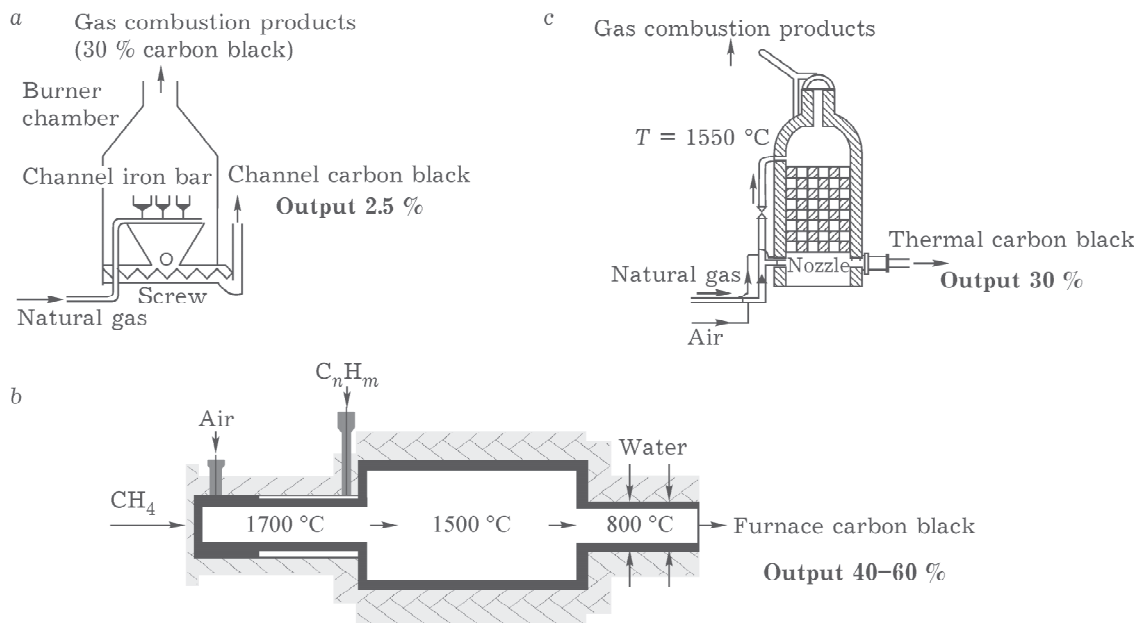
Fig. 2. Reactors for the synthesis of carbon black by different methods: *a* − thermal, *b* − furnace, *c* − channel.

ature reaches 1500−1600 °C; after that, the delivery of air and feedstock is stopped, the outlet pipe is closed, and the feedstock is delivered to the top of generators, where carbon black is synthesized at a specified temperature. Since this is a cyclic process, two generators are commonly mounted, after which an aerosol containing the produced carbon black is cooled in refrigerators. This is followed by a maximum possible separation of carbon black. The purified effluent is ejected to the atmosphere or added to the feedstock gas intended for the next cycle of carbon black synthesis. Although 50 % of the produced carbon black is retained by the packing and burns in a new cycle, the yield of carbon black manufactured by the thermal method is quite high and can reach 30 %. However, the resulting product has large primary particles with highly non-uniform sizes [14].

The furnace method is applied more widely and produces various grades of carbon black *via* thermal oxidative decomposition of liquid or gaseous feedstock. The furnace method (see Fig. 2, *b*) is implemented in special furnaces that have three main zones: the combustion zone where a temperature necessary for decomposition of initial gaseous or liquid feedstock is maintained, the reaction zone, and the quenching zone where the aerosol is cooled with water. Fuel gas and air are fed into tangentially

arranged burners and their combustion provides high temperatures. The dehydrated feedstock heated to 250−300 °C goes to the reaction zone. The feedstock is sputtered with high-pressure air preheated in a stove to 350−400 °C. When temperature in the reaction zone is in a range of 1200−1700 °C, the hydrocarbon feedstock decomposes with subsequent formation of carbon black. To terminate the gasification reaction, the mixture is cooled with water injected by mechanical atomizers. Cooling of the aerosol stream is performed in three steps: first with water to a temperature of 900−1000 °C, then in a common collector to 650−800 °C, and finally in a refrigerator-evaporator with water to 250 °C. Carbon black with widely varying properties can be produced with a yield of 40−60 % by controlling the synthesis conditions [13].

Until recently, carbon black was also produced by the channel method (see Fig. 2, *c*). The synthesis was performed from the gaseous raw materials in the burner chambers by depositing carbon black on the deposition surface located over the gas burners, and carbon black was formed in their flame. Thus produced carbon black had the highest oxidation state of the surface, but its yield was very low, ca. 2.5 %. In distinction to other methods, the channel method of carbon black synthesis implies a great amount of wastes ejected to the atmo-

sphere with off-gases, ca. 30 % of the total target product [13]. The production of channel carbon black was prohibited exactly because of its high emission to the environment. The last channel carbon black production in Russia operating at the Sosnogorsk gas processing plant was closed five years ago [2].

However, emission of carbon black to the atmosphere is inevitable also in the furnace process. Since such emission occurs in all cases, industrial carbon black plants are equipped with systems for the recovery of the dust-like product. The recovery systems consist of several sequentially mounted cyclones and filters, which make it possible to considerably reduce the emission of carbon black to the environment in aerosol stream.

In cyclones (Fig. 3, a), carbon black particles are deposited by centrifugal force. Dust-laden gases tangentially move to the top of cyclone at a rate of ca. 12 m/s, gyrate, then move downward over the conical part of cyclone, are thrown to the cyclone walls, and fall to the cyclone hopper by gravity. The purified gas forms an internal stream and is removed through the cyclone outlet pipe. A rotating scroll mounted in a cyclone-concentrator prevents a loss of carbon black at the bottom of cyclone hopper; the scroll is used to remove a part of gases together with the recovered carbon black to the gas transport system. The recovery efficiency of a cyclone-concentrator with the rotating scroll increases to 92 % in comparison with 70 % for a conventional cyclone. The concentration of carbon black at the cyclone inlet is up to 100 g/nm^3, and at the outlet, up to 10 g/nm^3 [14].

A system of filters is commonly mounted after the cyclones. Carbon black plants until recently applied the schemes with deposition of aerosol in electric filters, cyclones and a wet cleaning system including foam gas coolers, Venturi tubes and settling chambers of suspensions with a complicated circulating water supply, however, carbon black sequestration in bag filters is more efficient [15].

Bag filters (see Fig. 3, b) can differ in the number of sections, the number of bags in a section, their length, diameter, and type of fabric. The principle of their operation is based on the upward passage of aerosol stream over
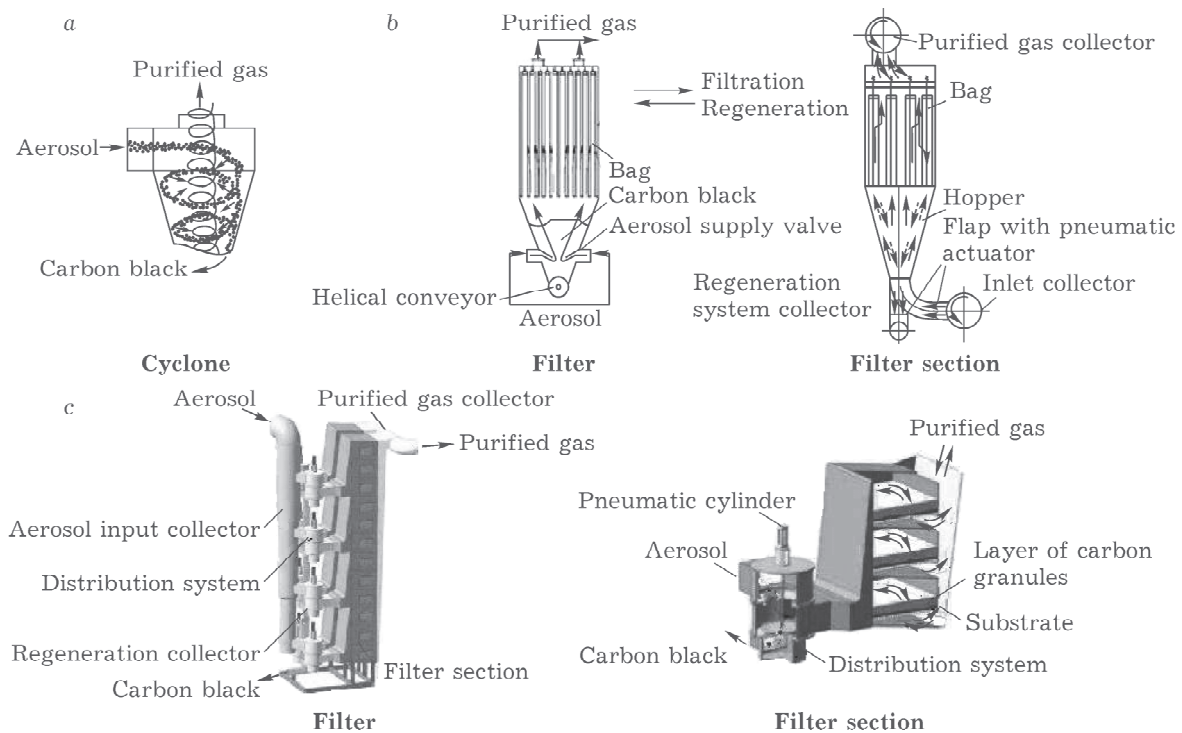

Fig. 3. Setups for the recovery of carbon black from aerosol streams: a − cyclone, b − bag filter, c − filter with a stationary bed of carbon granules.

the bag with deposition of the carbon black layer on the bag surface. The deposited layer is called AutoFilter, and a further efficient filtration of aerosol proceeds precisely through this layer, the bags are commonly regenerated by a reverse air blowing [14, 16].

The best bag filters equipped with bags of a glass fabric modified with polytetrafluoroethylene allow trapping carbon black with a particle size of 10 nm and greater. In addition, the application of such glass fabric extends the interrepair life of a bag filter [17]. However, such filters have some drawbacks, in particular, the necessity of frequent (every 6−24 months) manual replacement of the bags, which requires a shutdown of the entire section because failure of even a single bag decreases the trap efficiency of the section and produces a volley emission of carbon black.

The indicated drawback is absent in filters with a stationary layer of carbon granules (see Fig. 3, *c*), which can be used to recover the target product of carbon black production. Upon filtration in a layer of granules, the aerosol is divided into elementary streams, which flow along granules of the carbon layer, thus facilitating the confinement of carbon black. The layer of carbon granules is periodically regenerated *via* fluidization with a purified gas stream [18, 19].

The filter with a stationary layer of carbon granules FNS-8/8 was tested on a pilot plant at the Institute of Hydrocarbons Processing with deposition of active carbon black grades [14, 18]. The filter medium was represented by a layer of granulated carbon black with the granulometric composition of 0.7−1.8 mm, layer height of 200 mm and bulk density of 330 kg/m^3. The concentration of carbon black particles at the filter outlet was 60−100 mg/m^3.

Our pre-conceptual study on filtration of carbon black aerosols in filters with a stationary layer of carbon granules showed that this process had some advantages over filtrations in a conventional bag filter. At a high rate of aerosol filtering in a setup with preliminary deposition of carbon black from aerosol in a cyclone-concentrator and then in a filter with a stationary layer of carbon granules, the recovery efficiency can reach 99.9 %, and expenses are lower due to the absence of complicated replacement of a filter medium. In addition, volley emissions of carbon black to the atmosphere are absent [14]. The recovered carbon black goes to the systems of its granulation, where the collected dust-like product is pelletized. Most often this is performed in granulators, where carbon black is mixed with water and additives [13]. At this step of carbon black production, its emission to the environment with industrial wastewater may occur; therefore, it is necessary to install filters with activated slurry [9].

Carbon black that remains in aerosol stream after the recovery is burned up in special furnaces where its outlet concentration commonly does not exceed 4 mg/m^3 [8]. Such industrial off-gases are ejected to the atmosphere.

## BEHAVIOUR OF CARBON BLACK IN THE ATMOSPHERE

The main detrimental effect on the environment is exerted by emission of carbon black with industrial off-gases to the atmosphere where it can remain for quite a long time. Carbon black does not affect the ozone layer; however, it can cause some chronic diseases of respiratory system and cutaneous covering under a long-term exposure. In addition, according to Classification of the International Organization for Cancer Research (IARC), carbon black is considered as a potentially carcinogenic substance [9].

The maximum permissible concentration of carbon black in air is virtually the same in different countries, ca. 3.5−4 mg/m^3 in industrial off-gases [9]. It is believed that if the concentration of carbon black in air is far from maximum permissible values, it does not exert a significant harmful effect on the human body and environment as a whole.

In the atmosphere carbon black can interact with different active oxygen species. A possible source of such species is hydrogen peroxide, which in its turn can emerge upon interaction of water vapor with ozone by the following reaction [20]:

$$O_3 + H_2O \rightarrow H_2O_2 + O_2$$
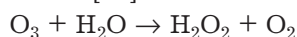
The main active oxygen species are various ions and radical ions that form from hydrogen peroxide by the scheme

$$H_2O_2 + H_2O \rightarrow HO_2^- + H_3O^-$$
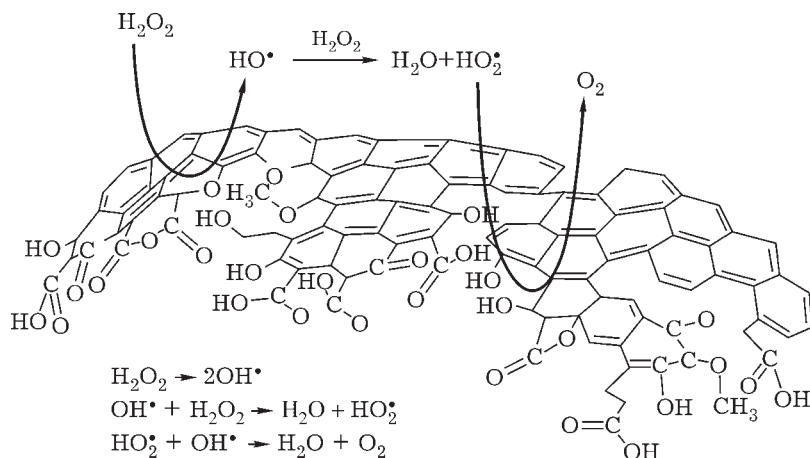$$O_3 + HO_2^- \rightarrow {}^\bullet HO_2^- + {}^\bullet O_3^- \quad [21]$$
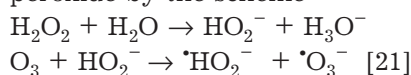
Fig. 4. Proposed scheme of H_2O_2 interaction with the carbon surface according to [22].

Interaction mechanisms of hydrogen peroxide with graphene layers of carbon black are reflected in the scheme (Fig. 4) [22].

According to this scheme, the hydrogen peroxide molecule is adsorbed on the surface and decomposes on active sites, which are represented by neutral or weak-acid functional groups. Intermediate surface products are desorbed and diffuse to the liquid phase where they can react with other hydrogen peroxide molecules to yield new intermediate products. Such intermediates diffuse to the carbon surface, interact with it and release molecular oxygen. The real surface of carbon black has oxygen-containing functional groups, the structure of which cyclically changes upon oxidation of carbon black [22, 23].

Adsorption of hydrogen peroxide molecules and the donor-acceptor interaction with electron transfer can proceed also on the basal plane of carbon black. This process results in decomposition of hydrogen peroxide by the radical mechanism similar to the Fenton reaction [24]:

$$AC + H_2O_2 \rightarrow AC^+ + OH^- + {}^{\bullet}OH$$
$$AC^+ + H_2O_2 \rightarrow AC + H^+ + {}^{\bullet}HO_2$$

Carbon black oxidation can proceed in two steps: functionalization and gasification. Gasification increases the concentration of defects in carbon layers and their roughness. As the roughness factor increases, so does the catalytic activity of carbon black in spontaneous decomposition of hydrogen peroxide [23].

The initial stages of carbon surface oxidation during functionalization lead to the forma-tion of phenol groups, which promote the spontaneous decomposition of hydrogen peroxide, and only then carboxyl groups are generated. The carboxyl groups shift pH of an aqueous suspension of carbon black to the acid region, thus inhibiting the hydrogen peroxide decomposition. Indeed, when carbon black is oxidized by hydrogen peroxide in an acid medium with ozone as the co-agent, pH of an aqueous suspension decreases much more rapidly, which is caused by the generation of carboxyl groups due to a decrease in the content of phenol groups [23].

Thus, carbon black particles emitted to the atmosphere are oxidized, intensively adsorb water vapor, and initiate condensation process in the clouds. A part of carbon particles settles due to coagulation and penetrates into soil and water areas.

BEHAVIOUR OF CARBON BLACK IN SOIL

Aerobic deterioration of carbon in soil, which is accompanied by a loss of adsorption activity, a decrease in pH and an increase in the O/C atomic ratio, proceeds in a wide range of operation temperatures, from −22 to 70 °C, commonly for 12 months [25], after that the process runs slowly with a loss in the particle weight for a period of 50 to 100 years or even longer [26]. The oxidized carbon reacts with inorganic soil substances, which inhibits its deterioration [27].

Carbon black can exert not only the detrimental but also the beneficial effect on soil by

catalyzing the destruction of organic compounds, particularly oil hydrocarbons [28, 29], and by improving the soil fertility [30].

Within the SB RAS complex integrated project No. 4.10 "New directions in the application of carbon black for the development of environmental materials and methods" (2006–2008), IHP of the SB RAS and the Institute of Petroleum Chemistry (IPC, SB RAS) performed a systematic experiment to assess the effect of carbon black on oil contaminated soils. The soil was contaminated with oil from the Sovetskiy oil field at a concentration of 70 g/kg and a soil to carbon black ratio of 100 : 2. The experiment was carried out at a temperature of 25 °C for 30 days in two variants: experiment and control (without carbon black). The population of hydrocarbon oxidizing microorganisms in soil was determined by their cultivation in solid nutrient media according to the standard microbiological technique [29, 31]. The population of ammonifier colonies was estimated after 6 days on a standard beef-extract agar, actinomycetes – after 7 and 15 days on a starch-ammonia agar, and fungus microflora – after 4 days on Czapek media (agars acidified with lactic acid) [31, 32].

Carbon black exerts the beneficial effect on soil and increases the population of all microorganisms under consideration [33]. The oscillating changes in their population are caused by the auto regulation mechanism (Fig. 5).

The population of ammonifiers, responsible for the destruction of oil hydrocarbons and decomposition of proteins, increases in 5 times in
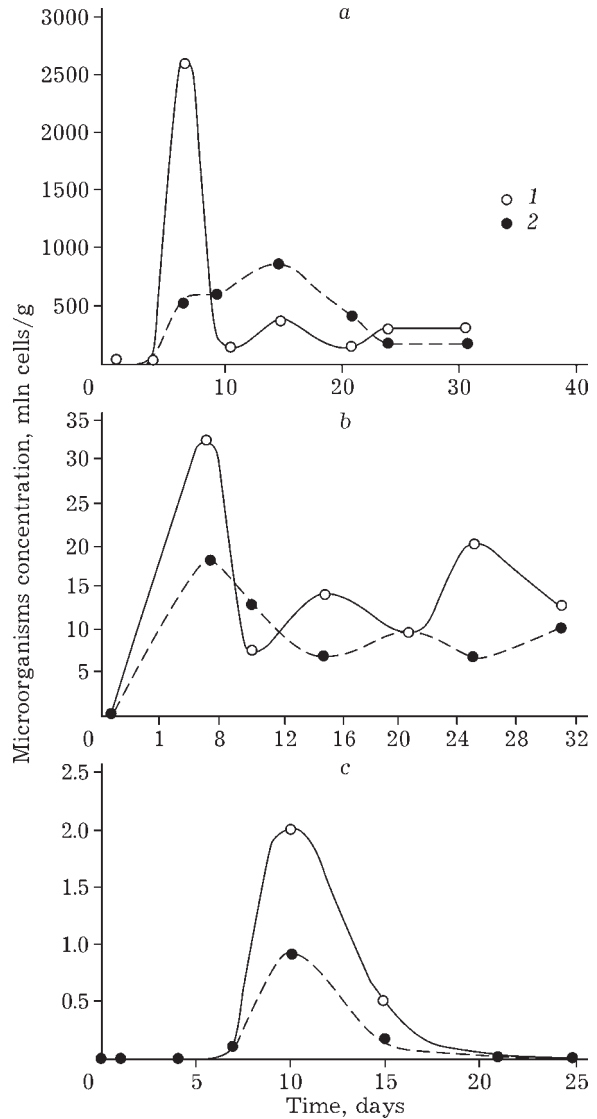
Fig. 5. Changes in the population of microorganisms on nutrient substrates with time: *a* – ammonifiers, *b* – actinomycetes, *c* – fungus microflora. *1* – with carbon black, *2* – without carbon black.
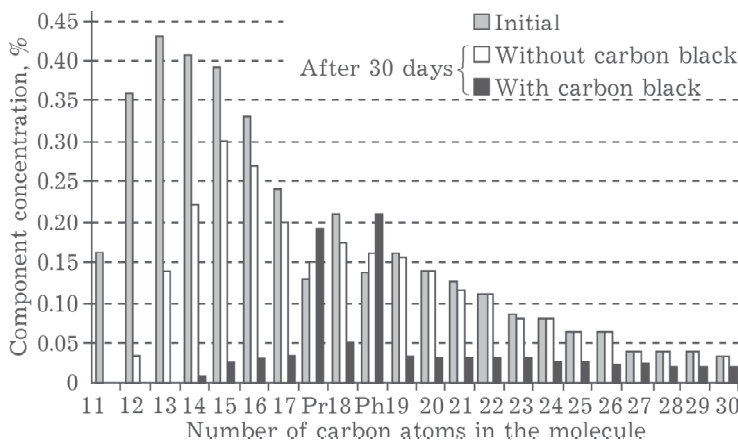
Fig. 6. Molecular mass distribution of hydrocarbons upon degradation of oil contaminants in soil [29].

the presence of carbon black. The population of actinomycetes, controlling the degradation of solid organic substance and accumulation of nitrogen in soil, increases in 2 times. The population of the fungal microflora in the soil with carbon black is increases in 2 times. The fungal microflora in the soil is responsible for nitrogen fixation and the soil fertility.

The initial and residual contents of saturated hydrocarbons after a 30 days exposure of soil microflora to oil contaminants were determined by gas-liquid chromatography [29]. The molecular mass distribution of hydrocarbons in the destruction process of oil contaminations in the soil changes in the presence of carbon black from $C_{11}-C_{30}$ toward $C_{14}-C_{30}$, moreover, their residual concentration is $2-3$ times lower than without carbon black (Fig. 6).

The biodestruction coefficient of oil in the soil is equal to the ratio sum of n-alkanes ($C_{17} + C_{18}$) to isoalkanes (pristane Pr + phytane Ph) [29, 32]. In the presence of carbon black, this coefficient is 4 times higher than without it. After a 30 day contact of the highly contaminated soil with carbon black, no more than 2 % of contamination remains in it [29].

The accelerated self-purification of soil from organic substances is caused by the adhesion of microbes on carbon black, which allows them to remain in the soil profile, and by the high sorption activity of carbon black toward both the oil products and the enzymes.

Carbon black can be involved in the respiratory function of soil microorganisms. Its functional groups (such as semiquinones) are electron donors for the reduction of oxygen molecule with the consecutive formation of various active oxygen species, in particular radical-ion superoxide and hydrogen peroxide, which are involved in the oxidative modification of hydrocarbons [34]. Enzymes of microorganisms belonging to the hydrocarbon oxidizing group additionally catalyze the reduction of oxygen molecule with the consecutive formation of various active oxygen species, thus accelerating natural self-purification of oil contaminations.

CONCLUSIONS

Carbon black is one of the varieties of black carbon having remarkable distinctive features in the structure, composition and properties. This is the product of the industrial synthesis manufactured in great quantities. Industrial emissions of carbon black can contaminate water areas, atmosphere and soil, and inevitable in its production.

Nevertheless, alongside with the traditional negative effect of carbon black, as a variety of black carbon, on health of living organisms and global warming a number of positive effects when it is injected into contaminated soil was discovered. Carbon black is recognized as a promising product for soil recultivation at their cleaning from hydrocarbons of oil and fertility enhancement.

The observance of safety rules to avoid emergencies, improvement and modification of existing installations of black carbon sequestration will allow minimizing the environmental pollution at its manufacturing.

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Основные научные аспекты эмиссии технического углерода

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Аннотация

Дано определение понятия "технический углерод" в соответствии с современными мировыми представлениями. Подробно описана структура технического углерода, морфология его агрегатов, элементный состав. В соответствии с международным стандартом показано место технического углерода среди других черных выбросов, имеющихся в атмосфере, кроме того, обрисованы принципиальные отличия технического углерода от сажи. Приведены источники эмиссии технического углерода в окружающую среду: атмосферу и почвы. Представлены промышленно реализованные способы получения технического углерода: термический, печной и канальный. Описаны принципиальные схемы установок улавливания технического углерода из аэрозольных потоков в процессе его производства, обеспечивающие максимально возможную эффективность очистки отходящих газов и сточных вод. Представлены схемы и принцип действия основных элементов систем улавливания технического углерода: циклонов и фильтров. Показано неблагоприятное воздействие технического углерода на окружающую среду в целом и на организм человека в частности, данные о предельно допустимых концентрация технического углерода. Описано поведение технического углерода в атмосфере и нефтезагрязненных почвах. Даны пути преобразования технического углерода в атмосфере, принципиальные схемы его окисления. Проведен ряд модельных экспериментов по исследованию влияния технического углерода на почвы, загрязненные нефтью. Выявлено положительное влияние технического углерода при очистке почв от нефтезагрязнений. Установлено, что технический углерод способен катализировать процессы деструкции различных органических соединений, в том числе углеводородов нефти, улучшая плодородие почв. Представлены сведения об аэробном старении технического углерода в почвах со временем.

Ключевые слова: технический углерод, эмиссия, окисление технического углерода, нефтезагрязнение почв, деструкция нефтезагрязнения