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Efficiency of Using Advanced Oxidation Processes and Activated Carbon for Municipal Wastewater Treatment

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Abstract—The global issue of hazardous persistent micropollutant accumulation in water bodies, including those of biological origin, was assessed. Modern approaches to municipal wastewater treatment using advanced biotechnological solutions (methods: SHARON, CANON, BABE, OLAND, ANAMMOX) and advanced oxidation processes (AOPs) were analyzed. Promising aspects of combining these methods to improve the removal of persistent forms of pollutants were identified. A methodology was developed for experimental studies on the removal of contaminants from aqueous solutions using ozonation, ultrasonic treatment, pressurized electrolysis, and the addition of powdered activated carbon as a reagent. The results of municipal wastewater treatment confirmed the potential of these methods. The best overall performance in terms of nitrogen compounds, organoleptic characteristics, and microbiological indicators was achieved by combining pressurized electrolysis with the addition of powdered activated carbon. This technological approach is proposed for use in the post-treatment of wastewater following biological treatment facilities.

Keywords: persistent biological pollutants, wastewater treatment, ozonation, ultrasound, electrolysis processes, powdered activated carbon, efficiency

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INTRODUCTION

Municipal wastewater treatment facilities in Russian and Belarusian cities do not fully ensure high-quality treatment of wastewater [1, 2]. The situation is further complicated by the development of the municipal and industrial sector, which leads to the formation of organic micropollutants, including novel highly persistent pollutants. These compounds occur at ultra-low concentrations and are extremely hazardous to living organisms, including humans [3]. The European Union has compiled a list of 432 substances that disrupt the human endocrine system and are prohibited in drinking water. In the United States, 41 million people receive drinking water containing xenobiotics of this class. The most frequently detected substances in municipal wastewater in various states include cotinine (92.5% of samples), cholesterol (90%), and carbamazepine (82.5%) [4].

As part of the effort to address this global sanitary, hygienic, and environmental challenge, the Stockholm Convention on Persistent Organic Pollutants adopted a document on May 22, 2001, which, among other things, calls for the following [5]:

— recognition that persistent organic pollutants are toxic, resistant to degradation, bioaccumulative, and subject to long-range transboundary transport through air, water, and migratory species; they are deposited far from the original source of emission and accumulate in terrestrial and aquatic ecosystems;

— acknowledgment of the need to take global action on persistent organic pollutants.

Accordingly, research on technologies for the reduction of persistent biological micropollutants in aqueous solutions, taking into account the inability of conventional systems operating at municipal wastewater treatment facilities to remove them, represents an important and highly relevant scientific and applied task.

REVIEW OF LITERATURE SOURCES

Persistent organic pollutants, including micropollutants, are organic compounds that are resistant to chemical, biological, and photolytic degradation and therefore remain intact for extended periods. They are characterized by [6] resistance to environmental factors, the ability to accumulate in biological organisms, potential for long-range transport, and a high likelihood of harmful effects on living organisms.

Among the advanced technological solutions for biological removal of pollutants from wastewater, which have potential for implementation at municipal wastewater treatment facilities of various configurations, the following methods are notable (some of them are still in the process of industrial adaptation and scale-up development):

— SHARON (Single Reactor System for High Activity Ammonium Removal Over Nitrite) is based on the use of a bioreactor in which ammonium nitrogen in wastewater is oxidized to nitrite, followed by the reduction of nitrite to nitrogen gas. The process relies on the difference in growth rates between ammonium-oxidizing bacteria (Nitrosomonas and Nitrosococcus) and nitrite-oxidizing bacteria (Nitrobacter) [7];

— CANON (Completely Autotrophic Nitrogen Removal Over Nitrite) is a process that also involves simultaneous nitrification and denitrification in a single reactor with limited oxygen supply. The key difference is that the denitrifying organisms act as anaerobic ammonium oxidizers, and highly concentrated aqueous solutions can be fed directly into CANON reactors [8];

— BABE (Bio-Augmentation Batch Enhanced) is a process that combines two technologies: nitrogen compound removal and augmentation of endogenous nitrifying organisms. The core of this method is the addition of nitrifying bacteria to the initial section of the treatment facilities, significantly reducing pollutant concentrations and the need for aeration. As a result, the biomass of activated sludge in the treatment facilities increases, which in turn enhances nitrification capacity throughout the system [9];

— OLAND (Oxygen-Limited Autotrophic Nitrification–Denitrification) is an autotrophic nitrification–denitrification process carried out under conditions of limited oxygen supply [10];

— ANAMMOX (Anaerobic Ammonium Oxidation) is the most promising method, involving the anaerobic oxidation of ammonium to molecular nitrogen using nitrite as the electron acceptor. The process takes place under anaerobic conditions across a wide temperature range. The final product is gaseous nitrogen, which can be easily removed from the solution [11].

A common limitation of the above biotechnological solutions is their insufficient oxidative potential for the effective removal of persistent organic pollutants, including micropollutants.

Meanwhile, modern advanced oxidation processes (AOPs) are based on oxidative mechanisms that involve the combined action of several factors [12]. These include Fenton reagent treatment, cavitation, photocatalytic oxidation, plasma chemical oxidation, the combined action of O_3 and H_2O_2, wet air oxidation, and supercritical oxidation.

Ozone, for instance, has a very high oxidative potential ($E_0 = +2.07$ V; in alkaline medium $E_0 = 1.24$ V) and decomposes organic compounds at standard temperature. During ozonation, multiple effects occur simultaneously: oxidation of contaminants, decolorization, deodorization, disinfection of wastewater, and oxygen saturation [13]. A number of successful implementations of ozonation systems have been reported in water supply and wastewater treatment infrastructure, including the WEDECON project (removal of micropollutants), the POSEIDON project (wastewater treatment in Braunschweig), the Swiss national project Strategy MicroPoll (Wüeri wastewater treatment plant in Regensdorf), and the PILOTOX project of the Technical University of Berlin (wastewater treatment at Berlin Ruhleben) [14–16].

Currently, significant progress is being made in research on the processes of indirect electrochemical oxidation of organic compounds using oxygen reduction intermediates under elevated water pressure [17, 18]. These processes involve the electrochemical generation of hydrogen peroxide from oxygen at the cathode, followed by chemical reactions between its intermediates and organic substrates in the electrolyte solution. In addition to enhanced generation of hydrogen peroxide due to the combined effect of pressure and electrolysis, there is also degradation of organic substrates by dissolved oxygen, further contributing to impurity oxidation [19].

Ultrasonic treatment of liquid leads to specific physical, chemical, and biological effects, including cavitation, capillary action, dispersion, emulsification, degassing, disinfection, local heating, and many other processes that promote the breakdown of organic components in aqueous solutions [20].

Of particular interest for the removal of biological micropollutants is a reagent-based technology recommended in Information and Technical Reference Document on Best Available Techniques (BAT) for Wastewater Treatment Using Centralized Sewerage Systems of Settlements and Urban Districts (ITS 10-2019). This approach, known as biosorption treatment, combines activated sludge processing with the use of powdered activated carbon. Hard-to-oxidize micropollutants are adsorbed on the surface of powdered activated carbon and subsequently oxidized.

Based on the expected potential for effective treatment of persistent micropollutants (as suggested in previous studies), the experimental investigations of organic pollutant removal from wastewater in this work employed ozonation, ultrasonic generation, pressurized electrolysis, the addition of powdered

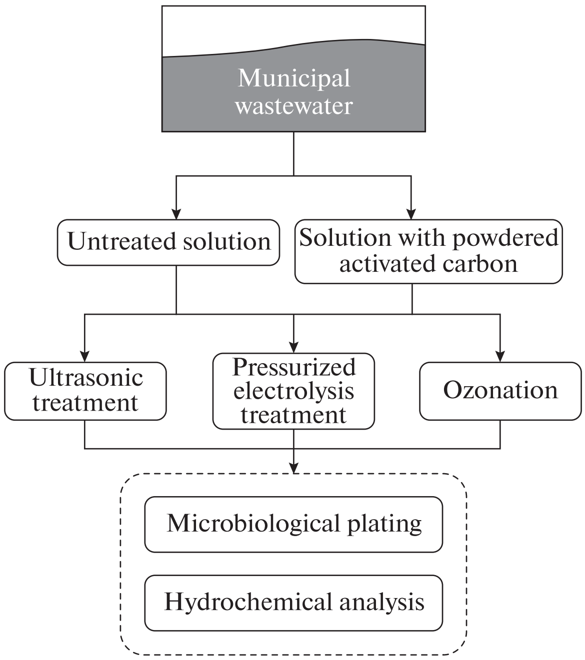

Fig. 1. Diagram of the experimental setup for evaluating the efficiency of using AOPs in municipal wastewater treatment.

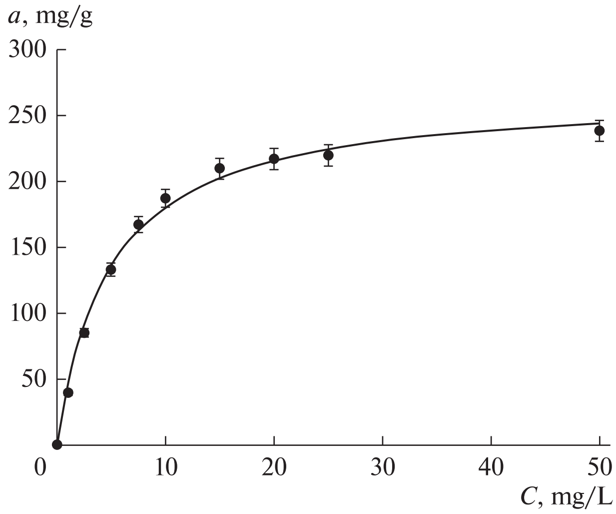

Fig. 2. Adsorption isotherm of methylene blue on EXTRASORB-101 activated carbon at 25°C.

activated carbon, and the combination of all non-reagent technologies with powdered activated carbon.

The aim of this work is to perform a comparative evaluation of the effectiveness of different AOP-based wastewater treatment processes, including ozonation, ultrasonic treatment, and pressurized electrolysis combined with adsorption on the surface of powdered activated carbon.

RESULTS AND DISCUSSION

Adsorption processes of solution components occur on the surface of the introduced activated carbon. The efficiency of the sorbent can be quantitatively characterized, for example, using the constants in the Langmuir equation. To determine these parameters, an adsorption isotherm (25°C) of methylene blue on EXTRASORB-101 granules was constructed (Fig. 2).

Langmuir equation

$$a = a_{\max} KC/(1 + KC), \quad (1)$$

where a is the adsorption, mg/g; $a_{\max}$ is the adsorption at maximum surface coverage of the sorbent, mg/g; K is the constant characterizing the adsorption–desorption equilibrium; and C is the equilibrium concentration of the adsorbate in solution. In linear coordinates, the equation can be written as (Fig. 3):

$$C/a = 1/(a_{\max} K) + C/a_{\max}. \quad (2)$$

From Fig. 3, the constants of the Langmuir equation were determined: $a_{\max}$ = 263.2 mg/g and K = 0.213. The calculated value of $a_{\max}$ indicates the high adsorption capacity of the activated carbon used. However, considering the high content of organic substances in the wastewater, the use of the sorbent alone is neither practical nor economically justified. It is more reasonable to use the sorbent in combination with physicochemical treatment of the wastewater in order to capture stable oxidation products of pollutants (Tables 1 and 2).

Ultrasonic (US) treatment is also proposed for water purification and degassing [22]. The first documented use of ultrasound for water treatment was reported in [23]. The authors described the occur-

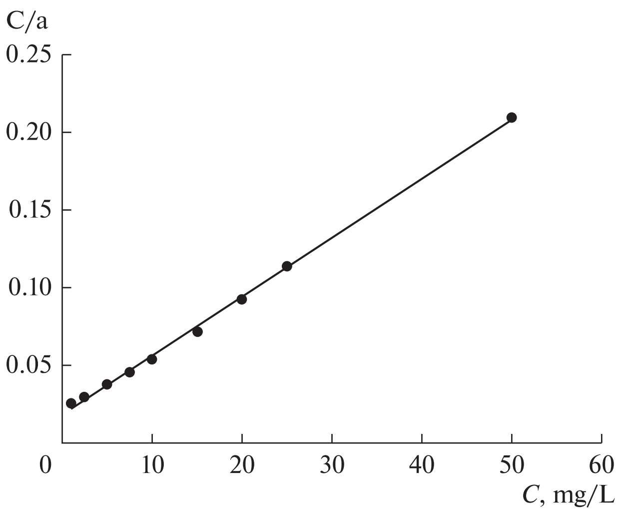

Fig. 3. Dependence $C/a = f(C)$.

Table 1. Results of AOP-based municipal wastewater treatment according to hydrochemical indicators

Wastewater treatment method	Results of hydrochemical analysis				
	pH	Redox potential, mV	$C_{(NH_4^+)}$, mg/L	$C_{(NO_2^-)}$, mg/L	$C_{(NO_3^-)}$, mg/L
Untreated municipal wastewater (before treatment)	9.04	–124	0.2	40	25
Wastewater + powdered activated carbon (1 g/L)	7.81	–46.2	0.15	0.2	15
Wastewater + O_3 (10 mg/L, 1 min)	8.08	–63.1	0.1	0.1	12.5
Wastewater + powdered activated carbon (1 g/L) + O_3 (10 mg/L, 1 min)	8.06	–64.8	0.1	0.1	12.5
Wastewater + ultrasound (2 W/cm^2, frequency 35 kHz, 2 min)	7.88	–51.6	0.1	0	14
Wastewater + powdered activated carbon (1 g/L) + ultrasound (2 W/cm^2, frequency 35 kHz, 2 min)	8.07	–62.1	0.1	0	14
Pressurized electrolysis (26 A, 48 V, 13 bar, exposure time 1.5 min)	7.94	–55.1	0.1	0.3	12.5
Wastewater + powdered activated carbon (1 g/L) + pressurized electrolysis (26 A, 48 V, 13 bar, exposure time 1.5 min)	7.91	–53.3	0.1	0.2	12.5

Table 2. Results of AOP-based municipal wastewater treatment according to organoleptic indicators

Wastewater treatment method	Organoleptic characteristics	
	light transmittance, %	odor, score
Untreated municipal wastewater (before treatment)	53.2	4
Wastewater + powdered activated carbon (1 g/L)	58.2	4
Wastewater + O_3 (10 mg/L, 1 min)	54.3	5
Wastewater + powdered activated carbon (1 g/L) + O_3 (10 mg/L, 1 min)	49.5	5
Wastewater + ultrasound (2 W/cm^2, frequency 35 kHz, 2 min)	55.4	4
Wastewater + powdered activated carbon (1 g/L) + ultrasound (2 W/cm^2, frequency 35 kHz, 2 min)	51.9	4
Pressurized electrolysis (26 A, 48 V, 13 bar, exposure time 1.5 min)	65.9	3
Wastewater + powdered activated carbon (1 g/L) + pressurized electrolysis (26 A, 48 V, 13 bar, exposure time 1.5 min)	72.1	3

rence of cavitation as well as the increase in the rates of chemical reactions in wastewater under ultrasonic exposure. The action of ultrasound on aqueous solutions can be attributed to cavitational, thermal, and chemical effects. The combination of all these effects explains the oxidative action of ultrasound on the components of aqueous solutions; however, the greatest effect is observed when ultrasound is combined with other treatment factors [24, 25].

As a result of ultrasonic exposure to aqueous solutions, free radicals are formed, initiating oxidation processes of the pollutants:

$$H_2O \xrightarrow{\text{ultrasound}} H^{\bullet} + OH^{\bullet},$$

$$H^{\bullet} + O_2 \xrightarrow{\text{ultrasound}} HOO^{\bullet},$$

$$HOO^{\bullet} + H^+ \rightarrow H_2O_2,$$

$$\text{Substrate} + OH^{\bullet} \xrightarrow{\text{oxidation}} \text{Oxidized form},$$

$$\text{Substrate} + HOO^{\bullet} \xrightarrow{\text{oxidation}} \text{Oxidized form},$$

$$\text{Substrate} + H_2O_2 \xrightarrow{\text{oxidation}} \text{Oxidized form}.$$

The results of AOP-based municipal wastewater treatment according to hydrochemical indicators are presented in Table 1. The results of evaluating the light transmittance and odor of the wastewater before and after treatment are shown in Table 2.

An analysis of the hydrochemical indicators of wastewater quality allows the following conclusions to be drawn:

— pH: the initial wastewater exhibited an alkaline medium (pH above 9), while after all types of treatment, it approached neutrality, with pH values around 8;

— redox potential: none of the treatment types caused significant fluctuations in the redox potential (recorded range: 65 mV to –47 mV, with an initial value of –124 mV);

— ammonium nitrogen concentration: a significant reduction (approximately 50%) was recorded for all treatment options, except for the use of powdered activated carbon alone, which achieved a 25% reduction;

— nitrite concentration: all treatment methods demonstrated high efficiency (initial concentration of 40 mg/L; results ranged from 0 to 0.3 mg/L);

— nitrate concentration: overall reduction was approximately 50%; the best results (12.5 mg/L) were achieved with pressurized electrolysis, the combination of ozonation and powdered activated carbon, and the combination of pressurized electrolysis and powdered activated carbon;

— solution light transmittance: the highest improvement in this parameter was observed with the synergy of pressurized electrolysis and powdered activated carbon (18.9%) and with pressurized electrolysis alone (12.7%). In contrast, the combinations of ultrasound and ozone with powdered activated carbon resulted in reduced light transmittance compared to untreated wastewater;

— organoleptic criterion (odor): the best deodorization was achieved through pressurized electrolysis and its combination with powdered activated carbon (1-point reduction), whereas ozonation alone increased the odor score by 1 point.

The results of the overall microbial contamination assessment of the wastewater are presented in Figs. 4–9.

In the initial solution (see Fig. 4), after streak plating, a relatively uniform distribution of colonies can be observed across the Petri dishes, with a denser concentration at the starting points of the streaks.

After ultrasonic treatment of the sample (see Fig. 5), a pattern similar to that of the initial solution is observed: the microorganisms are distributed evenly across the dishes, with an increase in the number of colonies near the starting points of the streaks.

The addition of powdered activated carbon results in a situation (see Fig. 6) that is nearly identical to that without its use, though a slight increase in the growth area of microorganisms on the nutrient medium is observed. It is also important to note that ultrasound may stimulate microbial activity and growth at low intensities and/or short exposure times.

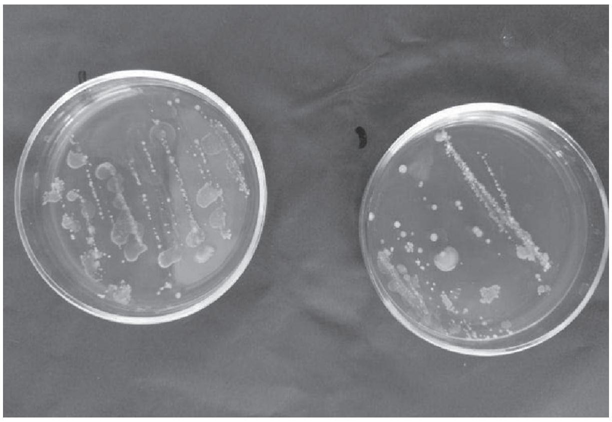

Fig. 4. Total microbial contamination of untreated municipal wastewater.

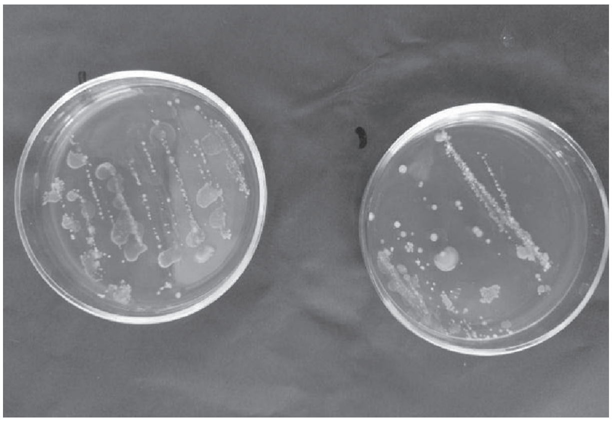

Fig. 5. Total microbial contamination of municipal wastewater after ultrasonic treatment.

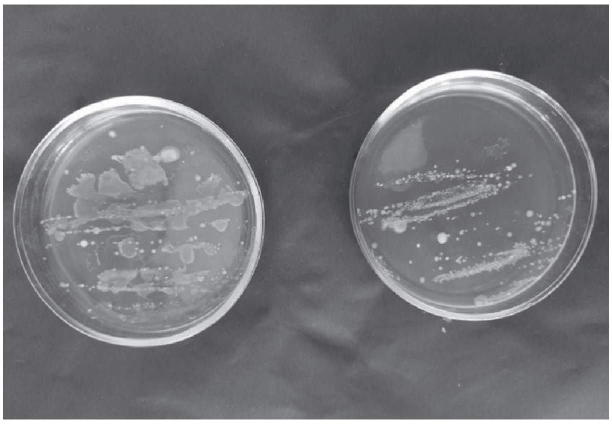

Fig. 6. Total microbial contamination of municipal wastewater after ultrasonic treatment with the addition of powdered activated carbon.

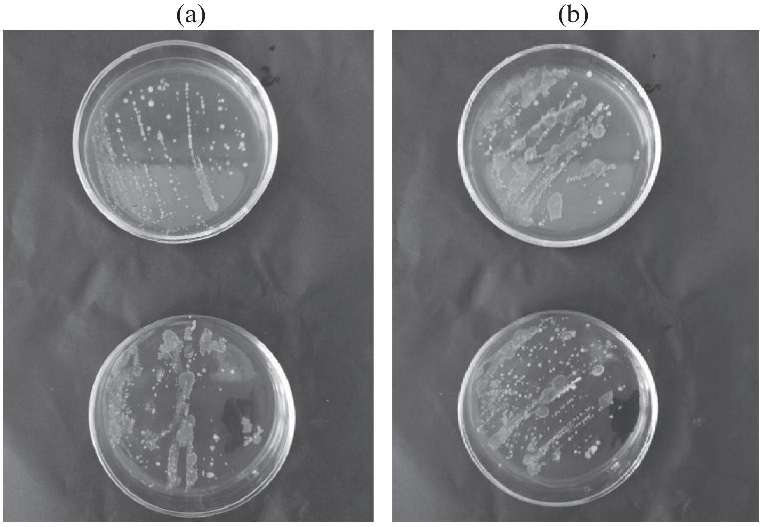

Fig. 7. Total microbial contamination of municipal wastewater after ozonation: (a) without powdered activated carbon, (b) with powdered activated carbon.

Ozone treatment did not result in a reduction in the total microbial count compared to the initial sample, both with and without the addition of powdered activated carbon (see Fig. 7).

A markedly different situation is observed in the sample treated with pressurized electrolysis in the presence of powdered activated carbon. As shown in Fig. 8, a decrease in the total microbial count is evident compared to all other samples, as confirmed by both replicates. A similar result is observed without the addition of activated carbon (see Fig. 9), although in one of the replicates there was significant microbial growth, possibly due to accidental contamination. The second replicate showed nearly complete sterility, with only a few colonies per Petri dish.

To assess the microbiological parameters of wastewater following conventional biological treatment (primary clarifier → aeration tank → secondary clarifier), a set of investigations similar to those above was performed (see Fig. 10).

In the sample of this treated wastewater (see Fig. 10), microorganisms continuously grew across the surface of the solid nutrient medium. This indicates that a substantial number of mesophilic aerobic and facultative anaerobic microorganisms remain in the aqueous solution after biological treatment, capable of rapid growth in the studied medium.

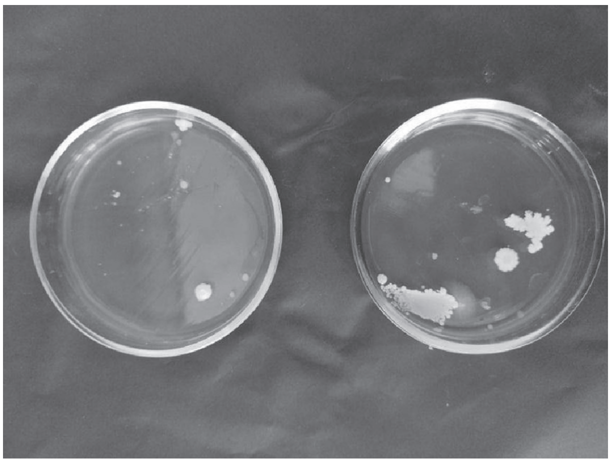

Fig. 8. Total microbial contamination of municipal wastewater treated with pressurized electrolysis with the addition of powdered activated carbon.

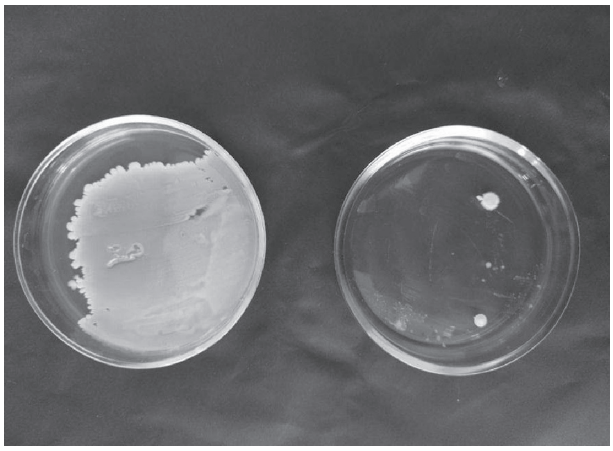

Fig. 9. Total microbial contamination of municipal wastewater treated with pressurized electrolysis without the addition of powdered activated carbon.

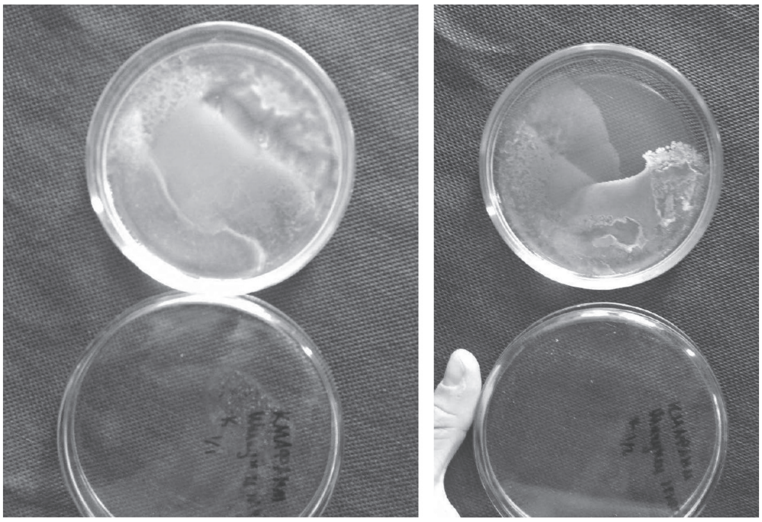

Fig. 10. Total microbial contamination of wastewater after conventional biological treatment.

CONCLUSION

The results of experimental investigations of wastewater treatment using various AOP-based technologies and their combinations with the dosing of powdered activated carbon allow the following conclusions to be drawn:

— the efficiency of the applied approaches (pressurized electrolysis, ozonation, ultrasonic treatment, and the use of powdered activated carbon as a reagent) for the reduction of nitrogen-containing compounds (ammonium nitrogen, nitrites, nitrates) reaches 50% and higher, which justifies their application for wastewater treatment (post-treatment);

— the highest performance in normalizing the organoleptic properties of wastewater is achieved through pressurized electrolysis in combination with powdered activated carbon (light transmittance, 18.9%; odor, approximately 25%).

The analysis of microbiological indicators of wastewater after treatment also confirms the highest efficiency of pressurized electrolysis with the addition of powdered activated carbon: a significant reduction in the total microbial count was recorded compared to the other approaches.

Thus, among the AOP-based technologies and their combinations with powdered activated carbon dosing, the most promising method is the approach based on electrochemical treatment of aqueous solutions under pressure with powdered activated carbon as the reagent. In this case, an almost complete reduction in the number of colony-forming bacterial units was observed, which may indicate the destruction not only of the bacterial cells themselves but also of their capsules and other dormant forms, the removal of which requires considerable resources. This is an indirect indication that such a combined method could also enable the elimination of persistent organic micropollutants when applied after conventional biological treatment.

Further research should focus on refining the operating parameters of electrochemical treatment under pressure with powdered activated carbon dosing and on developing industrial-scale systems and solutions.

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CONFLICT OF INTEREST

The authors of this work declare that they have no conflicts of interest.

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