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Purification of surface water by using the corona discharge method

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Abstract

Purpose. The primary objective of this research is to develop a technology for achieving water quality at the level of Maximum Permissible Concentrations (MPC) required by sanitary regulations and norms.

Methods. To meet these regulatory requirements, experimental studies were conducted to analyze the chemical and microbiological composition of water, focusing on parameters such as the total microbial count (quantity/ml), coliform bacteria (quantity/ml), coliphages (quantity/100 ml), clostridia (cl/20 ml), and other harmful substances. The research also examined water disinfection processes depending on the autumn and winter seasons of the year.

Findings. To assess the device's effectiveness, the research determined the optimal ozone concentration and contact time in the disinfection chamber. The findings indicate that 0.6 grams per hour (G = 0.6 g/hour) of ozone (O_3) per cubic meter of surface water is sufficient for the removal of harmful microbiological substances.

Originality. The primary innovation in this research lies in the establishment of parameters for an ozone generator utilizing a novel corona discharge method. The study introduces both theoretical frameworks and practical methodologies for effectively disinfecting surface waters using this innovative technique.

Practical implications. This case study offers insights that can be applied and replicated in regions facing similar water quality challenges.

Keywords: *corona discharge, ozone, primary water, reservoir, ozonator, ozonated water*

1. Introduction

In the context of the mining industry, the effective management of water resources is an important task. Water is crucial in mining, including ore processing, dust suppression, and equipment cooling. However, the utilization of water in mining activities often results in the generation of contaminated surface water, posing significant environmental challenges [1], [2]. The purification of surface water is therefore essential not only to meet regulatory standards but also to ensure sustainable mining practices [3]-[5]. In this regard, the utilization of innovative purification methods becomes very important for mitigating environmental impact and maintaining operational efficiency.

Water disinfection with ozone is one of the most efficient methods for water purifications [6]-[8]. In many countries, including the USA, EU, Asian countries, ozone technologies have been used for water purification since 1980. To this day, ozone purification technologies are still in constant development. The application of ozone has some specific issues related to its chemical characteristics [9]-[11]. Ozone is triatomic oxygen with a specific smell and color. Ozone molecules are very unstable, since ozone dissociation into oxygen molecules and atoms occurs within a few minutes, depending on environmental conditions [12], [13]. In such a short period of time, it quickly removes harmful substances contained in water, including total microbial count (quantity/ml), coli index (quantity/ml), coliphages (quantity/100ml), clostridia (quantity/20 ml) and other harmful microorganisms. Ozone strongly oxidizes heavy metals contained in water. Ozone is the strongest oxidizing agent, since hydroxyl or peroxide is involved in chemical reactions. The purification of water with the same concentrations of organic and inorganic substances and pathogenic bacteria requires more ozone than the equivalent of active chlorine. At the same time, ozone purification technologies are preferable in comparison to water disinfection with chlorine [14]-[16].

The virucidal effect when disinfecting with ozone is higher than when using chlorine technologies. In addition to the bactericidal effect, the ozonation process removes smell, unpleasant tastes and changes the color of the water [17]. Discoloration occurs due to oxidation of color compounds. Oxidized molecules break down into colorless simple molecules. If water is treated with excess ozone, the smell of the water will not change, since in a few minutes it decomposes and turns into oxygen. The technological process of using ozone in water purification involves obtaining the gas on site and then immediately introducing it into the purification procedure. The tools for setting up the ozonator consist of the following process steps:

– preliminary air preparation (filtration, drying and cooling);

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– synthesis of ozone from atmospheric air, pure oxygen preparation;

– the process of mixing the resulting air-mixed ozone gas with water.

To carry out this case study, tools have been developed to install a full-cycle water purification ozonator. A corona discharge-based ozonator has been constructed and tested to purify surface waters from the Vyacheslav Reservoir located near the city of Astana. Our team reviewed previous works on surface water purification with an emphasis on ozone application methodologies in comparison with other technologies before constructing a corona discharge-based ozonator for the city of Astana.

Lin et al. indicated that by-product concentrations can be in the high range for surface water purification and control using halogen disinfection products with ozone [18]. Humic acid was used as a model compound. Zhao et al. compared chlorine-generated disinfection through the product formation from sequential use of ozone and monochloramine with alternative disinfection. Four main oxidation stages were carried out to assess the formation of ozone/monochloramine, monochloramine, chlorine/mono-chloramine, chlorine decontamination by-products. It has been found that most of the residual chlorine decomposition occurs during the first day, and the pH of the solution should be monitored more closely during monochlorination to prevent the automatic decomposition of monochloramine [19]. Wu et al. focused on an automated-control direct ultrafiltration (UF) process for water treatment, demonstrating its effectiveness in providing clean drinking water from micro-polluted surface water in a Chinese mountain village without the need for pretreatment or chemical cleaning. The study highlighted the process's simplicity and cost-effectiveness, with an operation and maintenance cost, showcasing its potential for application in rural regions with similar water quality challenges [20]. Abdykadyrov et al. worked on water purification using the high-frequency ETRO-02 corona discharge-based ozonator. This work was carried out in order to purify surface water from impurities of petroleum products, phenol, and hydrogen sulfide. In their comparison studies, they considered that ozonating water has a greater advantage over chlorination [11]. Lei et al. investigated a multifunctional water purification system that utilizes a self-powered ozone generation approach based on a Triboelectric Nanogenerator (TENG) with a "like charges repel" strategy (LCR-TENG) [21]. The study demonstrated effective bio-sterilization in water, showcasing the system's potential for treating organic matter and microorganisms. The design also allows integration of various external loads, such as ultraviolet lamps and sensors, expanding the system's functionality without disrupting ozone generation.

Gere et al. investigated the impact of water treatment, specifically chemical disinfection using hypochlorite or hydrogen peroxide, on the therapeutic components of thermal waters in seven Hungarian spas with varying water compositions [22]. The study revealed that while both disinfectants improved microbial quality, hypochlorite significantly reduced therapeutic components such as sulfide, bromide, and iodide ions by 40-99%, leading to high levels of disinfection by-products. In contrast, hydrogen peroxide only affected sulfide ions with a 91% reduction. The findings emphasize the importance of selecting an appropriate treatment procedure based on water composition to achieve microbial safety without compromising the therapeutic qualities of thermal waters.

Vaju et al. worked on improving the quality of drinking water by physical methods using middle-frequency inverters [23]. They recommended the use of several oxidizers and various decontamination devices, for example, decontamination by radiation of O_3 + UV (in the first stage ozone and then UV radiation). Such a combination improves water quality substantially. The disadvantages of physical methods are related to the increase in electricity consumption. Researchers have proposed a more energy-efficient inverter for UV lamps. Wang and Liao proposed a planar UV reactor with five distinct UV-C LED arrangements for drinking water treatment, employing computational fluid dynamics and ray tracing methods to simulate velocity fields, microbial trajectories, and irradiance distributions. Among the arrangements, turn-enhanced emission, combined with turn-induced secondary flow, demonstrated the highest efficiency in sterilizing Escherichia coli due to synergies between hydrodynamics and UV radiation, resulting in a significant inactivation value of 4.56 log at a flow rate of 300 mL/min. This study highlights that optimizing LED arrangements and channel configurations can effectively enhance the sterilization efficiency of UV reactors [24]. Authors of the research [25] and [26] explored the applications of microplasma technology in water treatment. In [25], the researchers investigated the efficacy and safety of a solar-powered point-of-use water ozonation system using microplasma technology, demonstrating its ability to reduce microbial contamination in laboratory and real-world settings, with favorable perceptions from end-users in a village in western Kenya. In [26], the study focused on the mechanistic and kinetic aspects of diazinone degradation using microplasma, revealing the efficiency of the degradation process influenced by various plasma-generated species and identifying the significant roles of oxides of nitrogen in the degradation mechanism.

Yang et al. worked on the removal of dimethylamine by ozone combined with hydrogen peroxide. Chemical oxidation reactions in water depend on pH level, so dimethylamine removal by ozone combined with hydrogen peroxide (O_3/H_2O_2) worked better, according to the research [27]. Xu et al. conducted a comprehensive comparison study between sand filtration (SF) and ultrafiltration (UF) in drinking water treatment, revealing differences in their efficiencies for removing various organic compounds. The findings provide practical insights for decision-making in adopting SF or UF technology, considering factors such as organic foulant removal and disinfection by-product formation [28]. Györök et al. developed the adaptive ozone generator with an integrated microcontroller [29]. During the technological process, ozone concentrations should be adapted according to water quality. As water quality deteriorates, ozone concentrations should be increased. Upper ozone concentration can be obtained by amplifying pulsed power discharges to improve ozone formation characteristics. Authors of the research [30] and [31] investigated innovative aspects of ozone production and suppression in air discharges. In [30], the researchers proposed a method of nonheating ozone suppression by controlling plasma chemical kinetics through adjusting pulse duration and repetition frequency, demonstrating a breakthrough in diversifying plasma technology for various applications. In [31], the influence of gas temperature on ozone generation and decomposition in a pulse discharge was explored, revealing that ozone decomposition is primarily caused by momentary increases in gas temperature within microdischarge channels, highlighting crucial insights for optimizing ozone synthesis processes in heated ozone generators. Forés et al. [32] conducted a study evaluating the pathogen disinfection efficiency of electrochemical advanced oxidation, highlighting its potential as a sustainable technology for water reuse with low energy consumption. The research explored the disinfection of bacteria, bacterial spores, bacteriophages, viruses, and protozoa, employing logistic, exponential, and linear models to describe inactivation kinetics. While the electrochemical process demonstrated high efficiency for bacteria and bacteriophages, it showed limitations in disinfecting viruses, spores, and amoebas.

Authors of the research [33]-[35] investigated diverse aspects of water treatment technologies. In [33], the focus was on ozone electrodispersion technology for water disinfection, exploring the effects of chlorine and ozone, and presenting experimental studies on water treatment parameters with ozone through the electrodispersion method. Research [34] delved into virus elimination using high-voltage pulses, achieving 100% inactivation of bacteriophages and proposing a method for capturing viruses from air. Research [35] provided a comprehensive review on support materials for the immobilization of nano-photocatalysts in water treatment applications, highlighting the potential of various substrates such as carbonaceous materials, polymers, glass, and emerging materials like aerogel and metal foams. These studies contribute valuable insights to the advancement of sustainable water treatment technologies.

Xingbin et al. [36] and Yuan et al. [37] conducted studies addressing the challenges posed by Chironomus kiiensis larvae in drinking water treatment. In [36], the focus was on enhancing the removal of these larvae in conventional water treatment processes through pre-oxidation. The study evaluated the inactivation efficiency of chlorine, chlorine dioxide, and ozone on Chironomus kiiensis larvae under different conditions. Results indicated that pre-oxidation, followed by conventional water treatment processes, synergistically enhanced the removal of the inactivated organisms, emphasizing the importance of combining oxidant inactivation with conventional processes for securing drinking water quality. In [37], the research explored the effects of multi-frequency ultrasound on inactivating Chironomus kiiensis eggs in a drinking water treatment plant. The study investigated stress responses, viabilities, stress-related protein concentrations, and antioxidative enzyme activities in Chironomus eggs subjected to single-, dual-, and triple-frequency ultrasonic radiation.

Mitsugi et al. investigated the properties of soil treated with ozone generated by surface discharge [38]. Soil and surface water around the world are polluted by agricultural pesticides and nitrogen fertilizers. Ozone mitigates nitrogen nutrient and bacteria poisoning. Karaca et al. worked on ozone production using dielectric barrier discharge plasma for COVID-19 and microbial inactivation [39].

In response to the challenges posed by the location of clean drinking water sources at significant depths, exceeding 200 meters, scientists and researchers globally are increasingly directing their attention towards the application of innovative purification methods [40]-[43]. This shift in focus is driven by the need to address the unique challenges associated with accessing and treating water from surface water sources. Ozone-based methods are environmentally friendly technologies for eliminating harmful substances in air and water with a strong bactericidal and viral effect. Hand and Cusick [44] systematically examine the factors influencing electrochemical disinfection system performance, including treatment context, oxidant selection, and operating practices. Their comprehensive analysis spans diverse water treatment scenarios, evaluating oxidant demand and disinfectant dose requirements in drinking water, centralized wastewater, and distributed wastewater treatment contexts for both free chlorine- and hydroxyl-radical-based systems. Furthermore, the authors highlight promising applications and operational strategies for electrochemical disinfection while proposing essential reporting standards for future research in this evolving field. Eguchi et al. worked on Laser Thomson Scattering (LTS) diagnostics for streamer discharge [45]. LTS diagnostic is considered a reliable method for measuring electron temperature and density in plasma. One of the main issues is high electricity consumption of purification technologies. Pichel et al. worked on optimization of a photovoltaic-photochemical hybrid system (SOLWAT) to improve the energy efficiency of water purification systems [46]. The researchers used a hybrid solar water disinfection and energy generation system. Escherichia coli (E. coli) response and photovoltaic efficiency were investigated for surface water and natural sunlight. The time required for solar disinfection can be halved, increasing purification by placing water on the top of the PV module.

Puviyarasi et al. investigated the inline electrolytic disinfection system for water supply using a Programmable Logic Controller (PLC). The PLC was used for water quality monitoring with the technological process of removing microorganisms. Hypochlorous acid (HOCl) and hypochlorite ionic compounds (OCl) are used for water disinfection with PLC testing [47]. Momai et al. investigated the efficiency of photocatalytic disinfection of drinking water using fixed plates coated with TiO_2 and WO_3 [48]. The results showed high inactivation efficiency of bacterial cells during 240 minutes of radiation. The catalysts were reused over four continuous cycles, and the results proved high stability and good reuse performance. Skiba et al. worked on the one-pot synthesis of silver nanoparticles and composite materials for drinking water disinfection, which gave a good effect. This technology has been found to be effective in disinfecting Coli to varying degrees [49]. Smail Latifa and her colleagues conducted research on microbial disinfection using a barrier discharge ozone generator [50]. The purpose of their study was to determine the neutralizing effectiveness of the ozone generator using microbiological analysis. The experimental results provided an optimal solution for this research objective.

Some foreign scientific studies cite work on the removal of Escherichia coli in water using corona discharge. Water disinfection there shows that the results are mainly determined by energy density. During water purification, water permeability, pH value and concentration of hydrogen peroxide and ozone are also measured to assess the result. Transmission electron microscopy images of Escherichia coli show changes in intracellular morphology following plasma treatment [51]. Izdebski et al. investigated the purification of river water from microorganisms using corona discharge and ozonation. Ozone treatment can reduce the concentration of microorganisms and bacteria by several percent in 45 seconds [52].

To date, many oxidizing agents (chlorine, chlorine dioxide, ozone, UV) are used in the process of surface water purification and disinfection, of which the properties of ozone are higher (Fig. 1) [53].

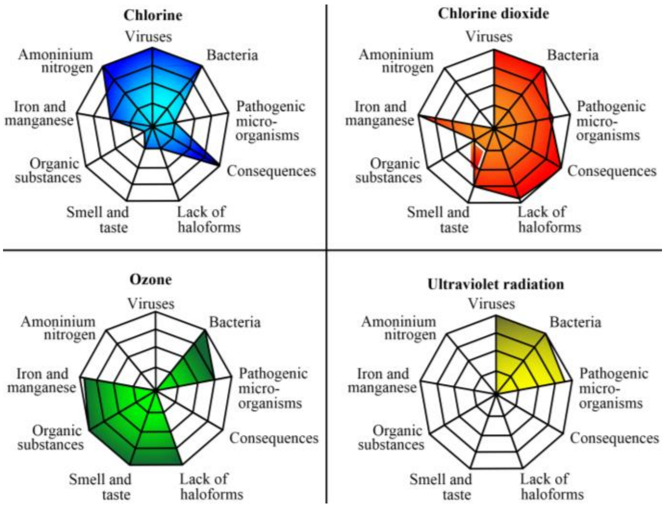


Figure 1. Efficiency of the surface water disinfection process

As shown in Figure 1, according to the tendency of destroying harmful microorganisms in water, high oxidizing properties of ozone can be observed. For this purpose, during the research work, a special pilot device has been developed – an ozonator based on the electric corona discharge.

Surface water ozonation is a widely used water purification process, particularly for disinfection and removal of organic and inorganic contaminants. Various factors influence water quality in the ozonation process, and it is important to consider both the benefits and potential challenges associated with this method. Figure 2 demonstrates advantages of the water ozonation process. Therefore, ozonation of surface water becomes a valuable method in water purification, offering numerous advantages, particularly in disinfection and elimination of diverse contaminants.

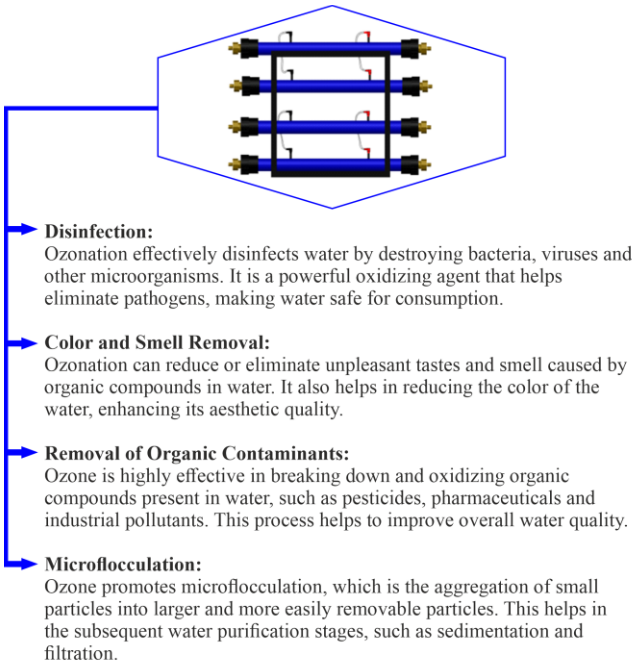


Figure 2. Advantages of the water ozonation process

However, effective management of this process is crucial to address potential challenges and guarantee the production of high-quality treated water.

This research uses experimental studies to analyze the chemical and microbiological composition of surface waters, investigates water disinfection processes in relation to seasonal variations, and introduces an innovative ozone generator using the corona discharge method. Ultimately, the main objective is to achieve water quality at the level of Maximum Permissible Concentrations (MPC) as required by sanitary regulations and norms.

2. Materials and methods

Currently, in ozonation technology, ozone synthesis by electric discharge is one of the main problems at the global level. Synthesized ozone can eliminate any harmful microorganisms contained in surface water and improve water quality. Water ozonation technology plays a pivotal role in addressing the growing global concerns about water quality and purification. Ozone synthesis through electric discharge is the cornerstone of this process, which aims to ensure efficient and sustainable treatment of water sources.

This paper provides a comprehensive overview of the current state of ozone synthesis by electric discharge, highlighting both advances and remaining challenges on a global scale. For example, after studying different water purification methods and technologies, we have developed and constructed our own electric corona discharge-based ozonator and tested surface water taken from the Vyacheslav Reservoir, located near the city of Astana. The reservoir area is 61 km², volume – 0.411 km³, width – 5.4 km, length – 11 km, average depth – 6.8 m, maximum depth – 25 m. The Vyacheslav Reservoir is used for irrigation and water supply to the city of Astana. Water mineralization level is 0.22-1.02 g/l. The Vyacheslav Reservoir water contains much harmful microbacteria, which are difficult to eliminate with existing water purification technologies. The municipal water supply organization "Astana Su Arnasy'' needs to prepare and supply 210 thousand/m^3 of drinking water for the residents of Astana city on a daily basis. Most of the water supply system networks in the city of Astana were built more than 50 years ago, which requires improvement of water purification facilities. Primary water is taken from the Yesil River (Fig. 3).

The pumping and filtering station processes surface water of the Vyacheslav Reservoir taken from the Yesil River (Fig. 3). The purification and treatment facilities at the station include 2 mixers, 20 settling tanks, 5 fast filters, 3 drinking and 1 technical water tank, 2 pumping stations, reagents, chlorination and other auxiliary facilities. The capacity of the pumping and filtering station is 210 thousand m^3/day. Surface water is first passed through the grids that trap debris, then treated with reagents at the water treatment plant, settled, after filtration disinfected to the required standards and stored in tanks (Fig. 3). Currently, the pumping and filtering station is processing water using 10 fast filters, including five filters with expanded clay and five filters with quartz sand and zeolite, which were taken from the Chankanai field located near the city of Astana.

To improve water quality at the facility (5), ozone-based water purification technological chain, such as an ozonator using corona discharge, has been developed (Fig. 4).

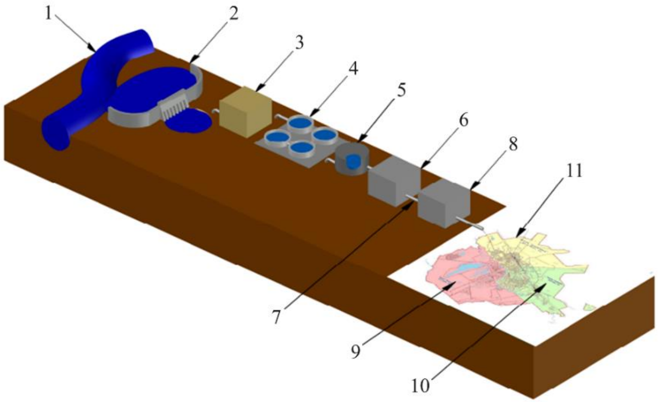


***Figure 3.** Water supply scheme of the city of Astana: 1 – Vyacheslav Reservoir, Yesil river; 2 – water zone; 3 – first wholesale water station; 4 – cleaning points; 5 – clean water reservoir; 6 – second wholesale water station; 7 – water transportation; 8 – third wholesale water station; 9 – water distribution for Saryarka district residents; 10 – water distribution for Esil district residents; 11 – water distribution for Almaty district residents*

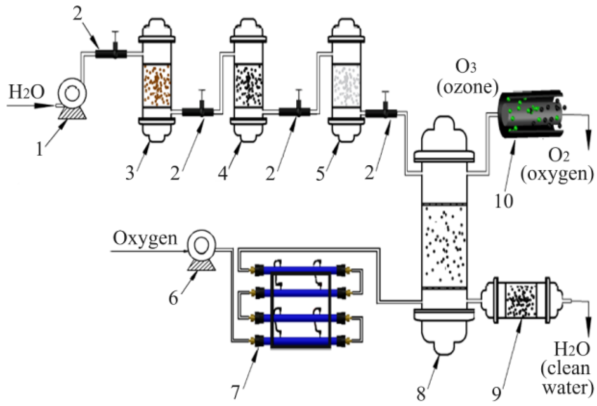


***Figure 4.** Water purification technological chain, ozonator using corona discharge for treating Vyacheslav Reservoir surface water taken from the Yesil River, near the city of Astana; 1 – pump with capacity of 10 m^5; 2 – valve (d = 36×40 mm); 3 – zeolite sand filter; 4 – activated carbon filter; 5 – quartz sand filter; 6 – air compressor; 7 – electric corona discharge-based ozonator; 8 – tank ($H_2O + O_3$); 9 – membrane filter; 10 – waste ozone decomposer*

Consistent monitoring of micro-bacteria was provided using MPC levels. The initial state of water quality for microbes, TCB (thermotolerant coliform bacteria), CCB (common coliform bacteria), coliphages and other components are shown in Table 1.

One of the main tasks of testing the ozonator is water purification to satisfy the state "Drinking Water" standard of the Republic of Kazakhstan #2874-82. Water purification should be intensified to eliminate the bacteriological components according to standards based on consistent monitoring.

***Table 1.** Chemical and microbiological composition of non-ozonated primary water of the Vyacheslav Reservoir, taken from the Yesil River near the city of Astana*

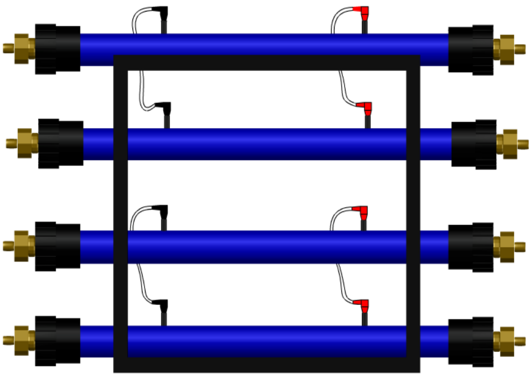
Parameters	Standards, not more (MPC)	Water sample characteristics, Autumn	Water sample characteristics, Winter
Transparency, mg/dm^3	2.00	4.13	4.58
Color, mg/l	20.0	5.0	5.0
Smell 20O U/m^3 (points)	2	0	0
Hydrogen index, pH	6.00-9.00	8.22	8.22
Permanganate oxidation, m nO_2 /dm^3	5.00	2.14	2.10
Ammonium salt, mg/dm^3	2.00 (N)	0.85	0.80
Copper, mg/dm^3	1.00	1.25	1.30
Nitrates, mg/dm^3	45.0	3.5	2.9
Nitrites, mg/dm^3	3.00	0.02	0.02
Total concentration, mole/m^3	7.0	4.2	4.2
Total iron, mg/dm^3	0.30	0.85	0.80
Sulfates, mg/dm^3	500.0	48.4	48.3
Dry residue, mg/dm^3	1000.0	311.5	313.6
Fluorine, mg/dm^3	1.20	0.31	0.30
Total number of microbes, 1 ml CFU (colony – forming units)	< 50	200	650
TCB (thermotolerant coliform bacteria) (100 ml CFU)	0	0	600
CCB (common coliform bacteria) (100 ml CFU)	0	350	700
Coliphages (100 ml Barrel of Oil equivalent BOE)	0	206	325

Coliphagesare are considered an indicator of enterovirus contamination, indicating that the water is contaminated with pathogenic microorganisms. The presence of parasitic pathogens is determined by the presence of Giardia Cysts in the water. Drinking water should be free of any pathogenic microorganisms.

The operation characteristics of the technological scheme are as follows: primary water, which does not meet the quality of drinking water, enters the sand load from the first zeolite *3* through a pump *1*. Water in the load made of this zeolite is pre-treated mechanically. Purified water passes through activated carbon, which adsorbs toxic substances *4*, and through a quartz filter, the water color changes and it comes into contact with ozone *8*. Ozone is supplied to the tank *8* through the ozonator *7* using a compressor *6*. After 30 minutes, the ozonated water is filtered through a membrane filter *9* and sent to consumers. Residual ozone deposited on the tank surface is decomposed using a destructor *10* and released into the atmosphere. The general image of an ozonator based on corona discharge *7* can be seen below (Fig. 5).

When ozone is dissolved in water, its concentration gradually increases, reaching the threshold value required for a given condition. The solubility of ozone in water depends on different conditions: water volume (V_{oz} / V_s), water temperature (T, °C) and pH value.

(a)



(b)

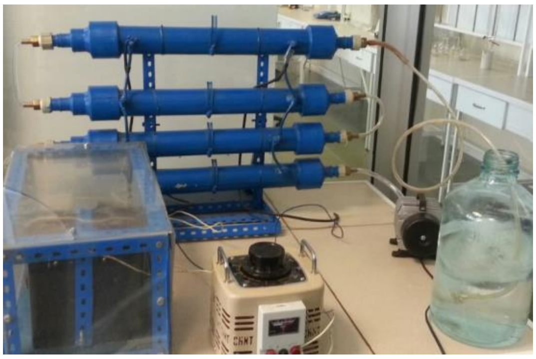


Figure 5. Electric corona discharge-based ozonator: (a) schematic representation of the ozonator; (b) image of the ozonator used for the study

The volume of dissolved ozone concentration (mg/l) in water is determined by the Bunsen coefficient. At the same time, the tendency of ozone to dissolve in water can be explained by Henry's law, according to which the pressure of dissolved ozone and gaseous ozone in solution are proportional. This pattern is written as:

$$C = \beta \cdot M \cdot P_{\gamma}\text{, g/l,} \tag{1}$$

where:

C – the solubility of ozone in water, g/l;

β – the Bunsen coefficient;

M – the density of ozone 2.14 g/l;

P_{γ} – the partial pressure of ozone in the melting gas medium [9].

The solubility of ozone in water is very high compared to oxygen and nitrogen in the atmosphere. Its solubility property in water increases with a decrease in water temperature (Table 2).

As can be seen from the experimental studies of foreign scientists (Table 2), water temperature (t, OS) increases and the Bunsen coefficient of ozone solubility in water (β) decreases. Ozone is less soluble in acidic and salt water than in distilled water. In research work, the change in Henry's constant influenced by pH at different temperatures can be expressed by the following ratios:

$$\ln H_a = 20.7 - \frac{3547}{T}\left(pH = 2\right); \tag{2}$$

$$\ln H_a = 18.1 - \frac{2876}{T}\left(pH = 7\right), \tag{3}$$

where:

H_a – the Henry's constant at the molar amount of ozone;

T – the absolute temperature, K. It can been observed that at a temperature of 300 K or 27°C, the solubility property of ozone in water is very poor.

Thus, it can be found that $\ln H_a = 8.9$ at pH = 2 and Ln $H_a = 8.5$ at pH = 7.

Table 2. Solubility property of ozone in water

T, °C	According to the checks in [54] Kogan B.F. Solubility Reference		According to the checks in [55] Masschelein W.J. Processus unitaires du traitement de L'eau potable		According to the checks in [56] Horvath M.L., Bilitrki Land Hutter. Ozone Ed. Akademia Kiado	
	β (l O_3 / l H_2O)	Solubility, g/l	β (l O_3 / l H_2O)	Solubility, g/l	β (l O_3 / l H_2O)	Solubility, g/l
0	–	1.13	0.51	1.09	0.64	1.37
5	–	–	–	–	0.50	1.07
10	0.41	0.87	0.38	0.78	0.39	0.83
15	–	–	–	–	0.31	0.66
20	0.32	0.69	0.29	0.57	0.24	0.51
25	–	–	–	–	0.19	0.41
30	0.26	0.56	0.21	0.40	0.15	0.32
35	–	–	–	–	0.12	0.25
40	0.21	0.45	0.15	0.27	–	–
50	0.17	0.37	0.10	0.19	–	–

The distribution of ozone in water as well as its dissolution in water at a particular point in time is also known from many scientific studies. As a result, research has shown that there is a hydroxyl – ion reaction in the zone concentration of ozone dissolved in water.

When ozone dissolves in water, it tends not only to react with chemicals present in the water, but also to its own distribution. These two processes occur simultaneously and depend on the temperature of the water, the pH of the medium, and the types of ions dissolved under ionic forces. In general, the ozone propagation rate in water can be written as:

$$\frac{-d\left[O_3\right]}{dt} = K_p \cdot \left[O_3\right], \tag{4}$$

where:

K_p – the constant of the ozone propagation rate in water.

A quantitative characteristic of the ozone tendency to dissolve in water can be determined as follows:

$$\begin{gathered} O_3 + H_2O \rightarrow 2\,OH^{*} + O_2O_3 + OH^{-} \rightarrow O_2^{-*} + HO_2O_3 + \\ + OH^{*} \rightarrow O_2 + HO_2^{*} \Leftrightarrow O_2^{-*} + H^{\pm}O_3 + HO_2^{*} \rightarrow 2\,O_2 + OH^{*}; \end{gathered} \tag{5}$$

$$2HO_2^* \rightarrow O_2 + H_2O_2. \quad (5')$$

This can be explained by the formation of the OH* radical and hydrogen oxide from the above equations. Ozone decomposes faster in an alkaline medium than in an acidic one, and in such conditions, the rate of its dissolution is expressed as follows:

$$\frac{-d[O_3]}{dt} = K_p[O_3] = K_a[OH-]\frac{1}{2}\cdot[O_3]^{3/2}, \quad (6)$$

where:

K_p and K_a – constants over a wide range of pH in water. The ionic strength of the stabilized phosphate buffer is 0.15 mole/m^3.

The presence and tendency to decompose of OH^- ions at or below pH = 3 is not significant. OH^- ions tend to dissolve faster in self-water in the range of pH = 7-10. Most often, at such pH values, the ozone propagation time in water is assumed to be about 10-25 min.

Currently, the Staehelin and Hoigne scheme takes into account all the main trends in ozone dissolution in water (Fig. 6).

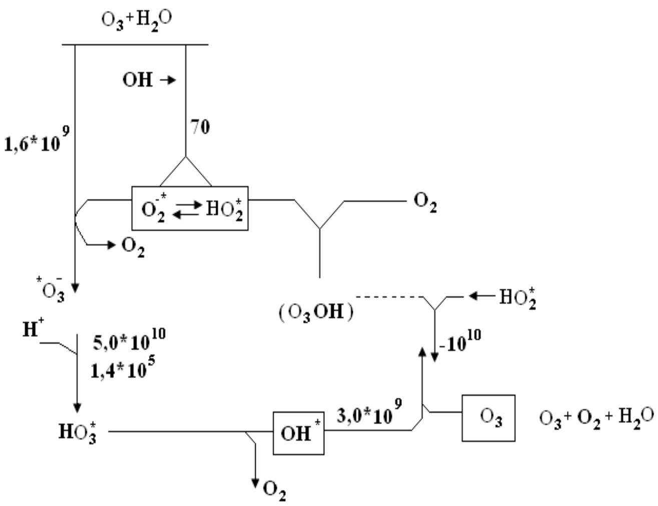


Figure 6. General cyclic scheme of ozone dissolution in water

Hydrogen oxide is able to accelerate the ozone self-decay reaction, giving rise to intermediate radical parts:

$$H_2O_2 \Leftrightarrow H^+ + HO_2^-; \quad (7)$$

$$HO_2^- + O_3 \rightarrow HO_2^* + O_3. \quad (8)$$

The equilibrium constant is 11.6.

Various solutions interact with ozone and the OH* radical, at which self-decomposition of ozone is accelerated. First of all, as a perfect radical, there are substances that cause oxygen in coal. For example:

$$HCOO^- + OH^* \rightarrow H_2O + COO^{-*} K = 3\cdot10^9 \text{mole}^{-1}\text{sec}^{-1}; \quad (9)$$

$$COO^{*-} + O_2 \rightarrow O_2^{-*} + CO_2. \quad (10)$$

Carbonate – bicarbonate ions, which remove OH* radicals from solution, are substances of the second group in the process of ozone decomposition in water. Understanding a completely accurate pattern of ozone dissolution in water, given the large number of water pollutants, is a difficult task. In the simplest case, half of the ozone is molecular, and the rest is converted into OH* radicals.

3. Results and discussion

Based on theoretical substantiation, experimental research was carried out with water sampling from the Vyacheslav Reservoir. When, under normal water purification conditions, the quality of purified water was brought up to the required standards, the concentration of viruses in it decreased by 80-93%. The better the purification of water from suspended inorganic and organic pollutants, the higher the efficiency of virus removal. In addition, during subsequent disinfection of water with chlorine, the presence of floating substances in it impairs the access of the oxidizing agent to bacteria and viruses. Therefore, the better the water is purified, the more effective the disinfection process will be. The types of bacterial contamination observed in the water source during the autumn and winter surveys included high total microbial counts (1 ml of CFU), CCB (100 ml of CFU), TCB (100 ml of CFU) and coliphages (100 ml of BOE) (Table 3).

In order to eliminate these above-mentioned microorganisms from the water, various amounts of ozone were released into the water and a purification process was carried out. The research results (Table 3) show the qualitative indicators of primary and purified water in terms of microbiological contamination (at ozone concentration of 0.6 mg/l).

In the table, according to microbiological indicators, the raw water does not correspond to the quality of drinking water. In it, the total number of microbes, CCB (100 ml of CFU) and TCB (100 ml of CFU) do not meet the indicators specified in the sanitary regulations and norms. In winter, it can be observed that the total number of microbes ranges within 650 – 50, and CCB – up to 2500 – 92. That is, microbiological indicators can be observed even in winter. During the technological process, special attention was paid to the elimination of microbiological pollutants when conducting research on purifying the water from the Ishim River using ozone. In Figure 7, we can observe the changes in microbiological indicators depending on the amount of ozone (time 30 minutes).

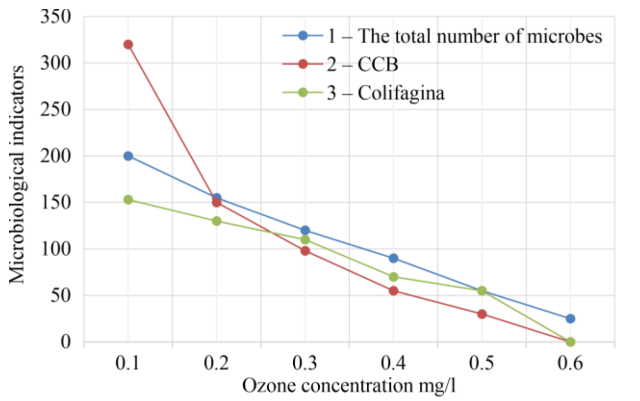


Figure 7. Relationship between microbiological indicators depending on ozone concentration (time 30 minutes): 1 – the total number of microbes; 2 – CCB; 3 – coliphages

Similarly, in Figure 8, a decrease can be observed in microbiological indicators depending on time (at this moment of time ozone concentration is 0.6 g/l). In the autumn-winter period, when treating raw and non – chlorinated water of the Ishim River with ozone in the amount of up to 0.1-0.6 g/l, the content of microbiological pollutants decreases by all indicators. And the most effective is the elimination of coliphages. The elimination efficiency of the total number of microbes is 60-70%.

Table 3. Disinfection of surface waters using corona discharge (microbiological composition of surface waters of the Vyacheslav Reservoir in autumn and winter)

Day	Total number of microbes (1 ml CFU)		CCB (100 ml CFU)		TCB (100 ml CFU)		Coliphages (100 ml BOE)	
	N*	V**	N*	V**	N*	V**	N*	V**
	200	45	320				153	
	150	40	360				120	
05.11.2021	150	30	240	destroyed	destroyed	destroyed	206	destroyed
	145	42	250				160	
	147	43	350				130	
	85	5	230				325	
	80	4	220				87	
15.12.2021	50	2	226	destroyed	destroyed	destroyed	80	destroyed
	65	1	200				60	
	60	0	220				75	
	78	4	400		350		156	
	65	3	700		450		140	
10.01.2022	60	2	500	destroyed	600	destroyed	135	destroyed
	55	1	350		350		155	
	60	3	360		300		150	
	500	47	92		325		150	
	450	44	110		500		135	
11.02.2022	650	45	120	destroyed	600	destroyed	120	destroyed
	560	45	110		400		120	
	350	35	120		550		100	

*Total number of microbes in water before initial ozonation
*Water content after ozonation (ozone concentration 0.6 mg/l)

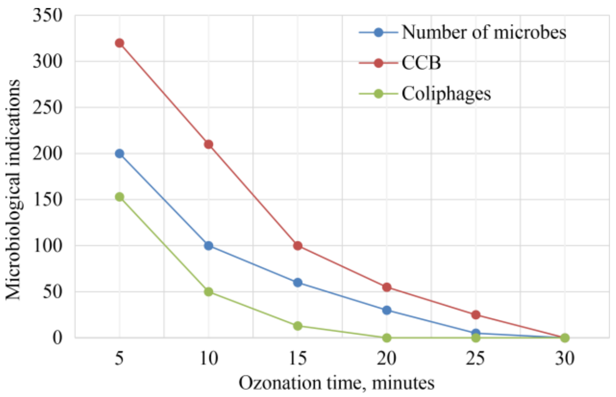


Figure 8. Influence of microbiological indicators on ozonation time at ozone concentration of 0.6 mg/l (T = 28°C pH = 7.5)

When comparing the effectiveness of water disinfection with ozone and chlorine, it is noted in foreign scientific studies [57]-[59] that water purification from microbiological pollutants largely depends on the amount of disinfecting agent. As indicated in the research works, it is impossible to purify and disinfect water using ozone alone. When using ozone and chlorine together, the required degree of water disinfection is achieved only when the chlorine dose is increased to 4 mg/l.

A special Mathcad program was used to determine the amount of ozone consumed per 1 m^3 of water in the process of ozone disinfection of reservoir water. The research algorithm is presented in Figure 9 below.

In Figure 9, $i = 1...n$ – the ordinal number of harmful substances contained in water; C_i – the number of harmful microorganisms in water after ozonation; X_i – the maximum permissible concentration of substances contained in water (MPC); b – constant value equal to 1; G_1, G_2, G_3, G_4 – the amount of ozone (g/hour); ϕ_0 – disinfected and purified water composition.

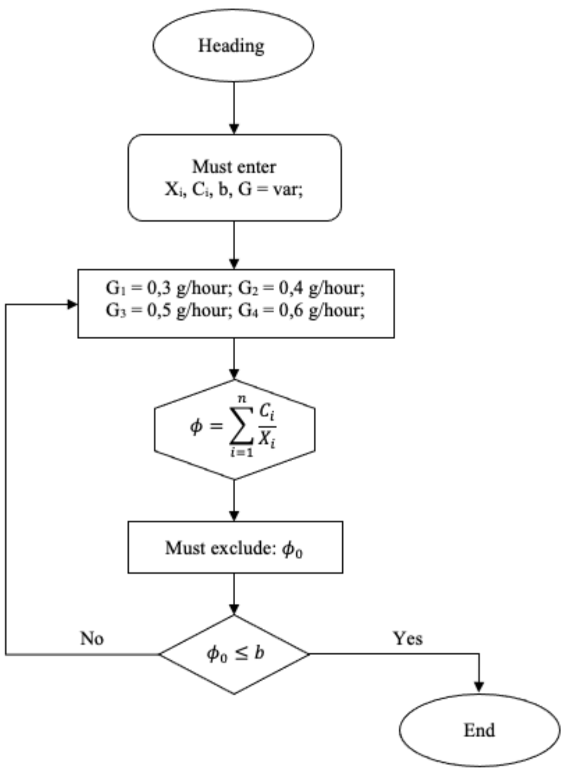


Figure 9. The process of reservoir water disinfection using ozone technology

Thus, the following parameters should be determined during the research:

– determining the tank volume;
– determining the time to fill the tank with water;
– release of different ozone amounts into water.

Before determining the total volume of water collected in the tank, the volume of water collected in small three tanks located inside the tank should first be individually determined.

To do this, we use the following Equations:

$$PV_1 = \left(a_1 \cdot b_1 \cdot h_1\right) + \frac{\pi l\left(R_1^2 + R_1^2 + R_1 r_2\right)}{3}, \text{ m}^3; \tag{11}$$

$$PV_2 = \left(a_2 \cdot b_2 \cdot h_2\right) + \frac{\pi l\left(R_1^2 + R_2^2 + R_1 r_2\right)}{3}, \text{ m}^3; \tag{12}$$

$$PV_3 = \left(a_3 \cdot b_3 \cdot h_3\right) + \frac{\pi l\left(R_1^2 + R_2^2 + R_1 r_2\right)}{3}, \text{ m}^3, \tag{13}$$

where:

$a_1 = a_2 = a_3$ – the tank length;

$b_1 = b_2 = b_3$ – the tank width;

$h_1 > h_2 > h_3$ – the level of water accumulated in the tank;

R_1 and r_2 – the large and small radii of the truncated cone located at the bottom of the tank;

l – the cone height.

To determine the volume of water collected in a common tank, we compact Equations (11)-(13) into a single Equation as:

$$PV_{total} = \sum_{i=1}^{3} PV_i \,. \tag{14}$$

In other way, the Equation (14) is described in general terms as follows:

$$PV_{total} = PV_1 + PV_2 + PV_3, \text{ m}^3, \tag{15}$$

where:

$PV_1 + PV_2 + PV_2$ – the volumes of small first, second and third tanks, respectively.

In order to determine the time spent on water filling these tanks, it is necessary first to determine the pump performance. If we assume that the pump performance in the unit is PQ = 10 m^3/hour, then the time spent for filling tank with water can be determined individually using the following Equations:

$$t_1 = \frac{t_{pump \cdot V_1}}{PQ}, \text{ min}; \tag{16}$$

$$t_2 = \frac{t_{pump \cdot V_2}}{PQ}, \text{ min}; \tag{17}$$

$$t_3 = \frac{t_{pump \cdot V_3}}{PQ}, \text{ min}, \tag{18}$$

where:

t_{pump} – the time to fill the tank with water using the pump of 10 m^3 (about 30 min);

V_1, V_2, V_3 – the volumes of the first, second and third tanks;

Q – the pump performance.

To determine the time for filling a total of three small tanks with water, we compact the Equations (16)-(18) into a single Equation as follows:

$$t_{total} = \sum_{i=1}^{3} t_i \,. \tag{19}$$

In other way, the Equation (19) is described in general terms as follows:

$$t_{total} = t_1 + t_2 + t_3, \text{ min}. \tag{20}$$

Since the water collected in the tank contains various chemical and bacteriological impurities, the summation effect is necessarily taken into account. Thus, it can be seen from Equation (21) that the sum of the real concentrations of chemical and bacteriological impurities contained in studied water should not exceed 1 in the ratio of C_1, C_2, $C_3 \ldots C_n$ to their MPC:

$$\frac{C_1}{MPC_1} + \frac{C_2}{MPC_2} + \ldots + \frac{C_n}{MPC_n} \le 1. \tag{21}$$

If the positive sides of Equation (21) become greater than 1, then measures to reduce the concentration of each substance should be considered. That is, it is necessary to consider the effective point of the amount of ozone released into the water composition. It is necessary to release different amounts of ozone into the water and check the result obtained by Equation (21).

In general, the above Equation (21) can be expressed using a mathematical equation such as:

$$F(x) = \sum_{i=1}^{3} \frac{C_i}{x_i} \le b\,, \tag{22}$$

where:

i =1-3 – ordinal number of substances contained in water;

$C_i = C_i + \ldots + C_3$ – the content of substances in water after ozonation;

$x_i = x_1 + \ldots + x_3$ – maximum permissible concentration of substances in water;

b – the constant 1.

During the technological process, it is possible to find out whether the purified water quality corresponds to Equation (2) or whether the water quality meets the standard.

For example, when conducting a full chemical and microbiological laboratory testing of primary water taken from the Vyacheslav Reservoir in winter, substances were detected that do not meet the maximum permissible concentration. The list of these substances is given in Table 4. And the algorithm of the disinfection process can be seen in Figure 9.

Table 4. Substances that do not comply with maximum permissible water concentration in the Vyacheslav Reservoir

No.	Names	MPC	Research results by season		Chemical and microbiological ozonated water composition, G (g/hour)			
			Autumn season	Winter season	0.3	0.4	0.5	0.6
1	Total microbial number, 1 ml CFU	< 50	200	650	120	90	55	25
2	TCB (100 ml CFU)	0	0	600	0	0	0	0
3	CCB (100 ml CFU),	0	320	700	98	55	30	0
4	Coliphages (100 ml BOE)	0	206	325	110	70	55	0
5	Transparency, mg/dm^3	2.0	200		200	650	120	0.09
6	Copper, mg/dm^3	1.0	0		0	600	0	0
7	Total iron, mg/dm^3	0.3	320		320	700	98	0.055

Depending on the amount of ozone, the quality of the disinfected water was tested individually using Equation (22). Check the quality of disinfected water according to Table 4 in general at an ozone concentration of 0.3 g/hour:

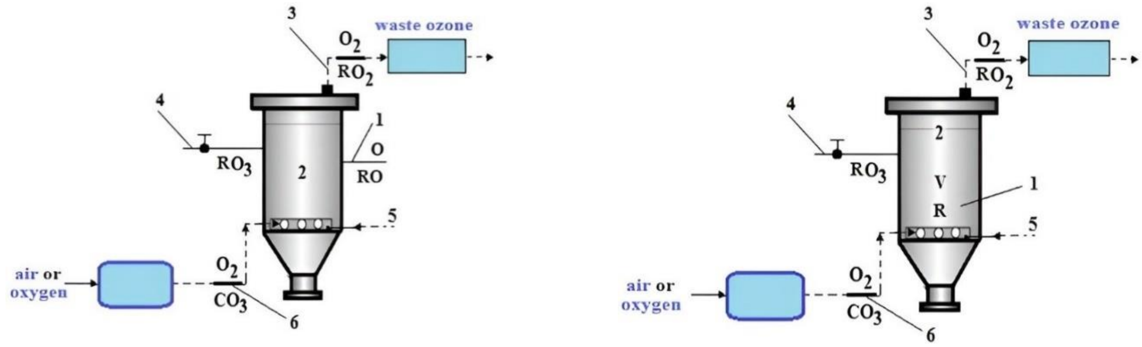
$$\frac{120}{50}+\frac{0}{0}+\frac{98}{0}+\frac{110}{2.0}+\frac{200}{1.0}+\frac{320}{0.3}\geq 1, \tag{23}$$

Equation (23) does not satisfy the condition given in Equation (21). This means that the water quality is at a lower level. Next, check the disinfected water quality at an ozone concentration of 0.6 g/h:

$$\frac{25}{50}+\frac{0}{0}+\frac{0}{0}+\frac{0}{0}+\frac{0.09}{2.0}+\frac{0}{1.0}+\frac{0.055}{0.3}\leq 1, \tag{24}$$

We see that Equation (24) satisfies the condition given in Equation (21). That is, it can be seen that the water quality meets the state standard 2874-82. The diagram of water ozonation process indicators in static and dynamic forms is shown in Figure 10.

There is a mutual similarity between static and dynamic modeling methods. The main difference between the water ozonation processes is as follows (Fig. 10).



$$G = G_O + G_R + G_B$$
$$G = [CO_3]\cdot Q_B$$
$$G_O = RO\cdot Q$$
$$G_R = [RO_3]\cdot Q_B$$
$$Q_B[CO_3] = RO\cdot Q + RO_3\cdot Q + [RO_3]\cdot Q_B$$
$$\eta = G - \frac{G_B}{G} = 1 - \frac{G_B}{G} = 1 - \frac{[RO_3]}{[CO_3]}$$
$$[CO_3]\cdot Q_B\frac{\eta}{Q} = RO + RO_3$$
$$D_{O3} = [CO_3]\cdot\frac{Q_B}{Q} = RO + \frac{RO_3}{\eta}$$
$$RO = \eta D_{O3} - RO_3$$

$$G = G_O + G_R + G_B$$
$$G = [CO_3]\cdot Q_B\cdot T_k$$
$$G_O = RO\cdot V$$
$$G_R = [RO_3]\cdot Q$$
$$Q_B[CO_3]\cdot T_k = RO\cdot V + RO_3\cdot V + [RO_3]\cdot Q_B\cdot T_k$$
$$\eta = G - \frac{G_B}{G} = 1 - \frac{G_B}{G} = 1 - \frac{[RO_3]}{[CO_3]}$$
$$T_k\cdot[CO_3]\cdot Q_B\frac{\eta}{V} = RO + RO_3$$
$$D_{O3} = T_k\cdot[CO_3]\cdot\frac{Q_B}{V} = RO + \frac{RO_3}{\eta}$$
$$RO = \eta D_{O3} - RO_3$$

Figure 10. Structural scheme of water ozonation process: a – dynamic mode; b – static mode; 1, 4 – primary and ozone-treated water; 2 – contact chamber; 3,6 – primary and exhaust ozone-air mixture; 5 – porous (porous) diffuser; G – amount of introduced ozone; G_O – ozone used to oxidize pollution; G_R – amount of ozone residue in water; G_B – amount of residual ozone in the gas mixture; Q – purified water consumption, m^3/h; V – volume of purified water, m^3/h; Q_B – consumption of ozone-air mixture, nm^3/h; RO – ozone absorption by primary water, mg/l; [CO_3] – ozone concentration in the post-ozonator impurity (ozone impurity air), G/nm^3; [RO_3] – ozone concentration in the ozone-air mixture after contact chamber, G/nm^3; η – efficiency of ozone use (efficiency); D_{O3} – amount of ozone, g/m^3; T_k – time of contact of the ozone-air mixture with water, min

The main error in the static simulation model is that the contact time in the contact chamber is not accurate because the residual ozone [RO_3] concentration in the gas is independent.

Since in this case there is a tendency towards saturation, water with ozone is of integral nature, the contact time cannot be determined by the formula $T_k = 60\,(V/Q)$, min (Q is measured in m^3/h, V – m^3). This formula is used in the course of dynamic practice. However, the supply of an ozone-air mixture from the ozonator to purified water at any given time does not take into account the parallel formation of the amount of ozone from the contact chamber and the spent ozone-air mixture [RO_3] = 0 (η = 1). In addition, in dynamic modeling, the value of η (efficiency) is not described in stationary dynamic mode [57].

As it turned out during the research, the most effective in most cases is a scheme using ozone and additional natural sorbents. Such technology provides for the elimination of organic and inorganic compounds by 80-85% in terms of oxidation, and by 96-100% in terms of biological indicators.

According to the research, the main problem today is insufficient study of surface water purification using the corona discharge method. Because today, thanks to the development of production centers, it is known to mankind that various toxic chemical compounds are highly polluting the natural waters of water bodies on Earth. As a result of the use of this contaminated water, various infectious and other diseases are transmitted through water today. Integrated research has been undertaken to study and discuss such issues.

For example, when studying and analyzing the water content of the Vyacheslav Reservoir by season, microbiological indicators that do not meet different maximum permissible concentrations (MPC) can be seen in the Tables 3 and 4. The Tables show that the total microbial count (1 ml CFU), TCB (100 ml CFU), CCB (100 ml CFU), coliphages (100 ml TCB), as well as the concentrations of heavy metals of copper (mg/dm^3) and total iron (mg/dm^3) do not meet the "drinking water" standard approved by the World Health Organization. The ozonator is used at various (0.3-0.6 g/hour) ozone concentrations for disinfection and purification of surface waters of the Vyacheslav Reservoir. The research on ozone concentration was carried out both theoretically and experimentally. As a result, 1m^3 of water is purified to the level of MPC and 0.6 g/hour of ozone is used for disinfection. This amount of ozone was found to be sufficient.

During the technological process, under the influence of ozone, bivalent iron (Fe^{2+}) is oxidized to trivalent (Fe^{3+}) iron. The trivalent iron ion precipitates in the form of hydroxide. As can be seen from Figure 10, the iron and copper concentrations in the water are 96-97%, and in terms of biological composition, microorganisms are destroyed by 96-100%. The main innovation of the research is the creation of an ozonator based on an electric discharge method according to a new model. In addition, experimental and theoretical studies have been performed.

4. Conclusions

Based on the results of research into the process of purifying surface water using the corona discharge method and ozone technology, a small laboratory ozonator model of a new type and a technological scheme have been developed at the Department of Electronics, Telecommunications and Space Technologies of the Kazakh National Research Technical University named after K.I. Satpayev for the disinfection of contaminated water of the Vyacheslav Reservoir. For practical testing of the unit, experimental and research work was carried out at the pumping and filtering station "Astana Su Arnasy". During the research, the following results have been obtained:

– microbiological indicators found in water, including thermotolerant coliform bacteria and microbes found in water in general, shows that it does not comply with various maximum permissible concentrations;

– it has been found that organoleptic indicators in the water composition changed the smell, taste, solubility of water and some heavy metals (iron and copper in general) oxidized and precipitated or some other chemical compounds;

– there has been a decrease in toxicological indicators found in water, i.e. inorganic compounds with a high toxicity index, such as nitrates, nitrites, fluorine, etc.;

– toxicological indicators were observed in the water composition, in which there was a decrease in organic compounds.

The research results have been obtained by examining different ozone amounts released into the water for conducting disinfection processes. Based on the research results, effective indicators of the water disinfection process in the Vyacheslav Reservoir have been determined. That is, 0.6 g of ozone is required to disinfect 1 m^3 of water. The disinfection time is 30 minutes. In the course of work to neutralize the microbiological bacteria found in the composition of water and to oxidize some chemical impurities, it has been found that ozone is a strong oxidizer. In addition, work on passing the disinfected water through natural filters was also conducted.

The above research has been performed directly with the use of an ozonator based on an electric corona discharge and made according to a new model. In the period from 2015 to 2022, research work was carried out and the main technological indicators of the installation were determined.

Author contributions

Conceptualization: AA, SA; Data curation: SM; Formal analysis: AA; Funding acquisition: AA; Investigation: AA, SA; Methodology: SA; Project administration: SA; Resources: YT; Software: YT; Supervision: AA; Validation: YT, KZ, SM; Visualization: KZ; Writing – original draft: AA, SA, YT, KZ, SM; Writing – review & editing: AA. All authors have read and agreed to the published version of the manuscript.

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Conflicts of interests

The authors declare no conflict of interest.

Data availability statement

The original contributions presented in the study are included in the article, further inquiries can be directed to the corresponding author.

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Дослідження очищення поверхневих вод коронним розрядом

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Мета. Основною метою наукової роботи був розгляд алгоритму робіт з доведення якості води до гранично допустимих концентрацій (ГДК), затверджених як санітарними правилами, так і нормами.

Методика. Для виконання затверджених вимог у ГДК проводилися різні експериментальні дослідження хімічного та мікробіологічного складу води. Прийнято ефективні рішення щодо усунення шкідливих мікробіологічних показників у складі води: загальної кількості мікробів (кількість/мл), індексу колі (кількість/мл), коліфагів (кількість/100 мл), клостридій (кл/20 мл) та інших шкідливих речовин. Роботи зі знезараження води вивчені стосовно осіннього та зимового сезонів року.

Результати. Для оцінки ефективності пристрою дослідженням визначено оптимальну концентрацію озону та час контакту у дезінфекційній камері. Оптимальна кількість озону для очищення 1 $м^3$ поверхневих вод становить 0.6 г на годину, G = 0.6 г/год (O_3), що достатньо для видалення шкідливих мікробіологічних речовин.

Наукова новизна полягає у встановленні параметрів роботи пристрою-озонатора з використанням нового методу коронного розряду. Дослідження представляє як теоретичні основи, так і практичну методологію ефективного знезараження поверхневих вод за допомогою цієї інноваційної методики.

Практична значимість. Це тематичне дослідження може бути застосовано та відтворено у багатьох інших регіонах з аналогічними водними проблемами.

Ключові слова: *коронний розряд, озон, первинна вода, водосховище, озонатор, озонована вода*

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