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Formation of reactive species during plasma treatment of wastewater from the mining and petroleum industries

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Abstract

Purpose. To investigate the regularities of reactive species formation during the plasma treatment of multicomponent wastewater from the mining and petroleum industries, as well as to assess their role in contaminant transformation and removal efficiency.

Methods. Laboratory experiments were carried out using a specially designed plasma-liquid reactor operating under high-frequency electrical discharge conditions (10-25 kV, 10-30 kHz, interelectrode gap 3-7 mm). The study included determination of the initial physicochemical characteristics of wastewater, including pH, electrical conductivity, total dissolved solids, and concentrations of heavy metals (Cu, Zn, Cd). During plasma treatment, the formation of reactive species (•OH, O_3, H_2O_2) was analyzed, and a kinetic model was applied to describe contaminant removal dynamics and treatment efficiency.

Findings. It was established that plasma treatment leads to the formation of hydroxyl radicals at a rate of $(1\text{-}5)\cdot10^{-6}$ mol·L^{-1}·s^{-1}, ozone in the concentration range of 10^{-6}-10^{-4} mol·L^{-1}, and hydrogen peroxide accumulation within 10-80 mg/L, thereby creating a pronounced oxidative environment. Copper concentration decreased from 20 to 0.5 mg/L (97.5%), zinc from 15 to 0.4 mg/L (97.3%), and cadmium from 0.5 to 0.02 mg/L (96.0%). The degree of organic contaminant degradation reached 70-90%. It was shown that the intensity of reactive species formation strongly depends on discharge parameters, while the proposed kinetic model adequately describes the experimentally observed treatment dynamics.

Originality. The study provides a comprehensive experimental and model-based analysis of reactive species formation during the plasma treatment of highly mineralized multicomponent wastewater from the mining and petroleum industries. Quantitative relationships were established among discharge parameters, reactive species generation, and contaminant removal efficiency.

Practical implications. The obtained results confirm the potential of plasma technologies for the advanced treatment of industrial wastewater with complex composition and high salinity. Practical implementation of the proposed approach may contribute to improved environmental safety, reduced reagent consumption, lower sludge generation, and expanded opportunities for water reuse in mining and petroleum production processes.

Keywords: *plasma treatment; wastewater; electrical discharge; reactive species; heavy metals; mining wastewater; petroleum wastewater*

1. Introduction

1.1. Problem statement

The mining industry has historically formed one of the core pillars of Kazakhstan's industrial development, substantially contributing to the formation and strengthening of the national mining and metallurgical complex [1], [2]. Today, this sector remains among the leading branches of the national economy, accounting for approximately 17-20% of total industrial output [3]-[5]. At the same time, rapid industrial growth together with the expansion of transport infrastructure has intensified anthropogenic pressure on natural ecosystems. The development of railway and road transport systems, as well as the modernization of energy and industrial facilities, has significantly increased environmental loads associated with emissions, energy consumption, and the exploitation of natural resources [6]-[10]. The active deve-lopment of the mining, oil, and gas sectors has led to the large-scale use of drilling, extraction, and reservoir exploitation technologies, which affect geological formations and subsurface water systems [11]-[14]. In addition, the expansion of industrial production and resource extraction requires increased water consumption and influences the state of surface and groundwater resources, which further intensifies environmental pressures on water ecosystems [15], [16].

Mineral extraction and processing operations, which rely heavily on water resources, generate substantial volumes of wastewater containing a wide range of contaminants. Environmental monitoring data indicate that large mining enter-

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prises produce approximately 20-40 million m^3 of industrial wastewater annually [17], [18].

Mine wastewater is generated during mining, mineral processing, and flotation operations and is characterized by elevated concentrations of heavy metals, dissolved salts, and organic compounds [18], [20]. Preliminary analyses showed that mine water contained copper at 10-25 mg/L, zinc at 8-18 mg/L, and cadmium at 0.2-0.7 mg/L, while total mineralization ranged from 1.5 to 3.0 g/L [20]. These values exceed the maximum permissible limits established by the World Health Organization and may result in aquatic ecosystem degradation and biodiversity loss [21]. Conceptual scheme of industrial wastewater generation in Kazakhstan's mining sector is shown on Figure 1.

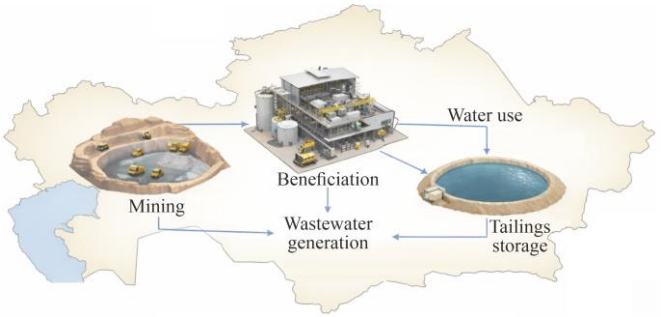


Figure 1. Conceptual scheme of industrial wastewater generation in Kazakhstan's mining sector

Although precipitation, coagulation, adsorption, and membrane separation are widely used for wastewater treatment, their reported performance remains limited [22]. According to the literature, physicochemical methods removed only 70-85% of heavy metals, whereas the degradation efficiency for persistent organic compounds remained at 40-60% [23]. In addition, the extensive use of chemical reagents and the generation of sludge reduced the environmental and economic feasibility of these treatment methods [24].

In this context, plasma technologies have emerged as one of the promising approaches for water treatment. Plasma treatment based on a high-frequency electrical discharge promotes the formation of highly reactive species in the liquid phase and at the plasma-liquid interface [25]. According to published and experimental data, plasma treatment generates hydroxyl radicals at rates of $(1\text{-}5)\cdot10^{-6}$ mol·L^{-1} s^{-1}, ozone at concentrations ranging from10^{-6} to 10^{-4} mol·L^{-1} and hydrogen peroxide accumulation within 10-80 mg/L [26], [27]. Owing to their oxidation potential of 2.07-2.80 V, these species create a strongly oxidative environment and facilitate the effective degradation of organic contaminants [28].

The efficiency of plasma treatment is governed by the intensity of reactive species generation, which in turn depends on the discharge parameters [29]. Preliminary experiments showed that the use of a high-frequency discharge operating at 10-25 kV and 10-30 kHz enhanced the formation of reactive species [30]. In addition, an interelectrode gap of 3-7 mm ensured stable discharge generation and maintained the stability of the plasma-liquid interface [31].

Nevertheless, the mechanisms of reactive species formation during plasma treatment of multicomponent systems with high ionic strength, such as mine wastewater, remain insufficiently understood [32]. In particular, quantitative relationships among discharge parameters, water composition, and the concentrations of generated reactive species are still limited, which hinders the industrial implementation of plasma technologies [33].

Therefore, investigating the formation of reactive species during the plasma treatment of mine wastewater represents an important scientific challenge. Such studies provide deeper insight into plasma-liquid interaction mechanisms, improve treatment efficiency, and support the development of environmentally safe water treatment technologies for the mining industry [34].

The main objective of this study is to identify the regularities governing the formation of reactive species generated during the plasma treatment of wastewater from the mining and petroleum industries and to assess their role in contaminant transformation.

To achieve this objective, the following tasks were defined:

– to determine the physicochemical characteristics of wastewater from the mining and petroleum industries;

– to experimentally investigate the formation of reactive species during plasma treatment;

– to analyze the relationship between discharge parameters and the intensity of reactive species formation;

– to assess the influence of reactive species on contaminant removal efficiency.

1.2. Review of current wastewater treatment methods

At present, physical, chemical, and biological methods are widely used for the treatment of industrial and mine wastewater. The literature indicates that, because of the complex composition of wastewater generated at mining enterprises, the use of a single treatment technology is often insufficient, and multistage treatment systems are therefore employed [35]-[37]. The efficiency of these treatment methods depends on the nature of the contaminants, their initial concentrations, and the operating parameters of the process.

Physical treatment methods are primarily aimed at removing suspended and dispersed particles. Studies have shown that mechanical filtration and sedimentation can remove 80-95% of particles in the size range of 10-1000 μm [38]. In membrane filtration, particle retention efficiency reaches 90-99% for particles in the range of 0.01-0.1 μm [38]. However, the removal of dissolved heavy metals and organic compounds remains limited to 20-40%, highlighting the inherent limitations of physical treatment methods [40]. The filtration process is characterized by the relationship among water flow rate, filter resistance, and pressure drop:

$$Q = \frac{\pi r^4}{8\mu L} \quad (1)$$

where:

Q – volumetric flow rate, m^3/s;

r – pipe radius, m;

μ – dynamic viscosity of the liquid, Pa·s;

L – pipe length, m;

π – constant pi (≈ 3,14);

ΔP– pressure drop across the filter, kPa.

During the experiments, the pressure drop varied from 10 to 50 kPa, while the filter resistance, depending on the material properties, ranged from 10^9 to 10^{11} m^{-1}.

Chemical treatment methods were used for the precipitation of heavy metals and the degradation of organic compounds. During coagulation and flocculation, the addition of

aluminum- and iron-based coagulants at concentrations of 50-150 mg/L resulted in the removal of up to 70-90% of copper, zinc, and iron ions [41], [42]. Ozonation and peroxide oxidation processes provided 60-85% degradation of organic compounds [43]. The kinetics of chemical treatment can be described by the following Equation:

$$\ln = \frac{C_0}{C_t}, \quad (2)$$

where:

C_0 – initial concentration;

C_t – concentration at time t;

k – reaction rate or inactivation constant;

D – applied dose or intensity parameter;

t – treatment time.

The reaction efficiency coefficient ranged from 0.02 to 0.12 min^{-1}. In addition, the amount of sludge generated during treatment was approximately 2-8 kg/m^3, which required further handling and disposal [44].

Biological treatment methods play an important role in the removal of organic contaminants. In activated sludge systems, biochemical oxygen demand (BOD) reduction reached 75-95% [45], [46]. In aerobic bioreactors, the rate of organic carbon degradation ranged from 0.3 to 1.2 day^{-1}, while biomass concentration stabilized at 2-4 g/L [46]. Biological treatment processes can be described by the following equation representing microbial growth kinetics:

$$\mu = \mu_{\max} \frac{S}{K_s + S} e^{-k_i C_i}, \quad (3)$$

where:

μ – specific microbial growth rate;

μ_{max} – maximum growth rate;

S – substrate concentration;

K_s – half-saturation constant;

k_i – inhibition coefficient;

C_i – concentration of inhibitory components, including heavy metals and dissolved mineral salts.

Experimental data showed that the microbial growth rate ranged from 0.4 to 1.0 day^{-1}, while the biomass yield was within 0.4-0.7 g/g. However, mine water mineralization at the level of 1.5-3.0 g/L and the presence of heavy metals reduced microbial activity, thereby limiting the overall treatment efficiency. The analysis showed that, despite the combined application of conventional treatment methods, complete wastewater purification was not achieved. Physical methods were effective for the removal of solid particles but showed limited performance with respect to dissolved contaminants. Chemical methods provided high removal efficiency; however, they were associated with significant reagent consumption and sludge generation. Biological methods effectively degraded organic matter, but their performance declined because of the presence of heavy metals at concentrations of 5-25 mg/L. Overall, Table 1 presents a comparative analysis of conventional wastewater treatment methods.

In recent years, advanced oxidation processes, particularly plasma-based technologies, have increasingly been considered as alternative treatment solutions. During plasma treatment using a high-frequency electrical discharge, the rate of hydroxyl radical generation was reported to be $(1\text{-}5)\cdot10^{-6}$ $mol\cdot L^{-1}\cdot s^{-1}$, while ozone concentrations ranged from 10^{-6} to 10^{-4} $mol\cdot L^{-1}$ and hydrogen peroxide accumulated to levels of 10-80 mg/L [25], [47]. The oxidation potential of these reactive species has been reported in the range of 2.07-2.80 V, enabling the degradation of up to 85-98% of organic contaminants [48].

Table 1. Comparative analysis of conventional wastewater treatment methods

Treatment method	Main target contaminants	Performance indicators	Process parameters	Main limitations
Physical methods	Suspended and dispersed particles	Removal of 80-95% of particles sized 10-1000 μm; membrane retention of 90-99% for particles sized 0.01-0.1 μm	Pressure drop: 10-50 kPa; filter resistance: 10^9-10^{11} m^{-1}	Removal of dissolved heavy metals and organic compounds limited to 20-40%
Chemical methods	Cu, Zn, and Fe ions; organic compounds	Heavy metal removal: 70-90%; oxidation of organic compounds: 60-85%	Coagulant dosage: 50-150 mg/L; sludge generation: 2-8 kg/m^3	Reagent consumption and the need for further sludge handling
Biological methods	Organic matter (BOD, organic carbon)	BOD reduction: 75-95%; organic carbon degradation rate: 0.3-1.2 day^{-1}	Biomass concentration: 2-4 g/L; growth rate: 0.4-1.0 day^{-1}; yield 0.4-0.7 g/g	Efficiency decreases at mineralization levels of 1.5-3.0 g/L and heavy metal concentrations of 5-25 mg/L

Thus, conventional treatment methods can be regarded as insufficient for the complete removal of the complex contaminant load characteristic of mine wastewater. This justifies the need for plasma technologies capable of creating a strong oxidative environment without the use of chemical reagents. Plasma treatment can therefore be considered a promising scientific and technological approach for the integrated treatment of wastewater generated by the mining industry.

1.3. Features of electrical discharge application in water treatment processes

Electrical discharge technologies for water treatment are based on plasma-chemical processes occurring under the influence of a high-frequency electric field. During discharge development, high-energy electrons, ions, and excited molecules interact with water components, leading to the formation of reactive species such as hydroxyl radicals (•OH), ozone (O_3), hydrogen peroxide (H_2O_2) and reactive nitrogen species [49]-[51]. These species possess high oxidation potential and initiate chain reactions that promote the degradation of both organic and inorganic contaminants. The main reactions responsible for the formation of reactive species (•OH, O_3, H_2O_2) are as follows:

$$e^- + H_2O \rightarrow H\bullet + \bullet OH + e^-; \quad (4)$$

$$H_2O^+ + H_2O \rightarrow H_3O^+ + \bullet OH; \quad (5)$$

$$O_3 + H_2O \rightarrow \bullet OH + O_2 + HO_2\bullet; \quad (6)$$

$$H_2O_2 + h\nu \rightarrow 2\bullet OH; \quad (7)$$

$$\bullet OH + \bullet OH \rightarrow H_2O_2; \quad (8)$$

$$HO_2\bullet + HO_2\bullet \rightarrow H_2O_2 + O_2; \quad (9)$$

$$O_3 + HO_2\bullet \rightarrow \bullet OH + 2O_2; \quad (10)$$

$$e^- + O_2 \rightarrow 2O\bullet + e^-; \quad (11)$$

$$O\bullet + O_2 + M \rightarrow O_3 + M; \quad (12)$$

$$O_2^- + O_2 \rightarrow O_3^- + O. \quad (13)$$

The plasma-liquid interaction zone plays a major role in contaminant removal, since electron impact reactions, ultraviolet radiation, and microshock-wave effects occur simultaneously in this region. Experimental studies have shown that plasma treatment resulted in the degradation of 85-98% of organic compounds and the removal of more than 90% of dissolved heavy metals through oxidation, precipitation, and complexation mechanisms [25], [52]. This combined effect is one of the key features that distinguishes electrical discharge technology from conventional treatment methods.

In addition to its high treatment efficiency, the electrical discharge method is characterized by environmental safety. Because no chemical reagents are introduced into the process, secondary pollution associated with reagent residues is avoided, while sludge formation is substantially reduced. According to the literature, the amount of sludge generated during plasma treatment was 3-5 times lower than that produced by coagulation-based methods, while the quality of the treated water met the requirements for industrial reuse [53], [54]. This feature enhances the environmental sustainability of plasma-based treatment technologies.

Another important advantage of electrical discharge technology is its energy efficiency. In plasma treatment, energy consumption depends on discharge voltage, frequency, electrode configuration, and treatment duration. Previous studies have reported energy consumption values in the range of 0.4-0.9 kWh/m^{-3}, which is competitive with other advanced oxidation processes such as ozonation and UV/H_2O_2 treatment [55]-[57]. Optimization of the discharge parameters made it possible to reduce energy consumption while maintaining treatment efficiency, thus demonstrating the practical potential of the technology. Figure 2 illustrates the relationship between electrical discharge parameter optimization and specific energy consumption during plasma water treatment.

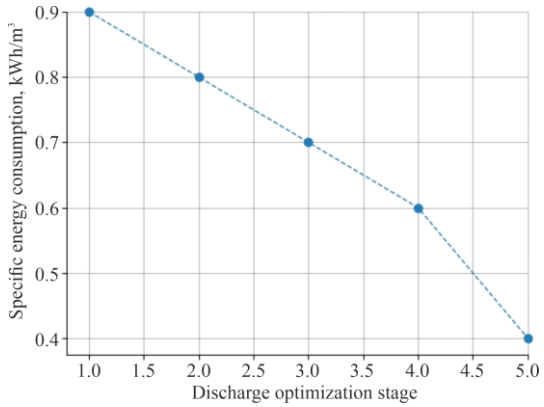


Figure 2. Reduction in specific energy consumption during discharge parameter optimization

Figure 2 shows a gradual decrease in specific energy consumption as the electrical discharge parameters are optimized for plasma water treatment. The results indicate that improved discharge stability enhances the utilization of reactive species, leading to higher treatment efficiency at lower energy input.

The versatility of the electrical discharge method broadens its field of application. Plasma processes have been effectively used for the removal of dyes, phenols, petroleum products, pharmaceutical residues, and heavy metal ions in various aqueous systems [57]-[60]. This indicates the potential for adapting electrical discharge technology to the treatment of mine wastewater characterized by high mineralization and a complex multicomponent composition.

A comparative analysis of the efficiency and environmental characteristics of electrical discharge technology is presented in Table 2.

Table 2. Quantitative performance indicators of water treatment by electrical discharge

Parameter	Unit	Range
Hydroxyl radical generation rate	mol·L^{-1}·s^{-1}	(1-5) 10^{-6}
Ozone concentration	mol·L^{-1}	10^{-6}-10^{-4}
Hydrogen peroxide accumulation	mg/L	10-80
Efficiency of organic contaminant degradation	%	85-98
Efficiency of heavy metal removal	%	90-97
Specific energy consumption	kWh/m^3	0.4-0.9
Energy density	kWh/m^3	0.3-1.2
Electron density	cm^{-3}	10^{12}-10^{14}
Treatment time	min	5-30
Discharge voltage	kV	10-25
Discharge frequency	kHz	10-30
Interelectrode distance	mm	3-7
Volume of water treated in the reactor	mL	200-300
Treatment temperature	°C	20-40
Sludge formation (relative)	times	3-5 times lower
Change in energy consumption at 85 → 95% efficiency	kWh/m^3	0.8 → 0.5
Oxidation potential	V	2.07-2.80

The relationship between treatment efficiency and energy consumption was identified as an important parameter for the industrial implementation of the technology. Experimental and modeling data showed that increasing treatment efficiency from approximately 85 to 95% reduced the specific energy consumption from 0.8 to 0.5 kWh/m^3. This effect was attributed to improved discharge stability and more efficient utilization of the generated reactive species.

Overall, electrical discharge technology for water treatment combines high efficiency, environmental safety, and energy efficiency, which makes it a promising method for the advanced treatment of mine wastewater. At the same time, establishing quantitative relationships among discharge parameters, reactive species formation, and contaminant transformation kinetics in multicomponent mine wastewater systems remains an important research need and is addressed in the following sections of this study.

1.4. Characteristics of wastewater from the Kumkol oilfield and the treatment approaches applied

The Kumkol oilfield, discovered in 1984, is one of the largest oil and gas fields in Kazakhstan and is located in the South Turgay Basin of the Kyzylorda Region. Oil production, processing, and associated industrial operations have resulted in the generation of substantial volumes of produced water and industrial wastewater containing hydrocarbons, dissolved mineral salts, suspended solids, and heavy metals

[61]-[63]. The multicomponent and complex composition of wastewater from the Kumkol field necessitates the use of effective treatment technologies.

Figure 3a and 3b illustrate the industrial and water treatment infrastructure of the Kumkol oilfield. Figure 3a presents an aerial view of the water treatment facilities, including storage tanks, settling units, and pipeline networks. Figure 3b shows a schematic representation of the oil production and processing infrastructure, including wells, separation units, collection systems, and produced water storage areas.

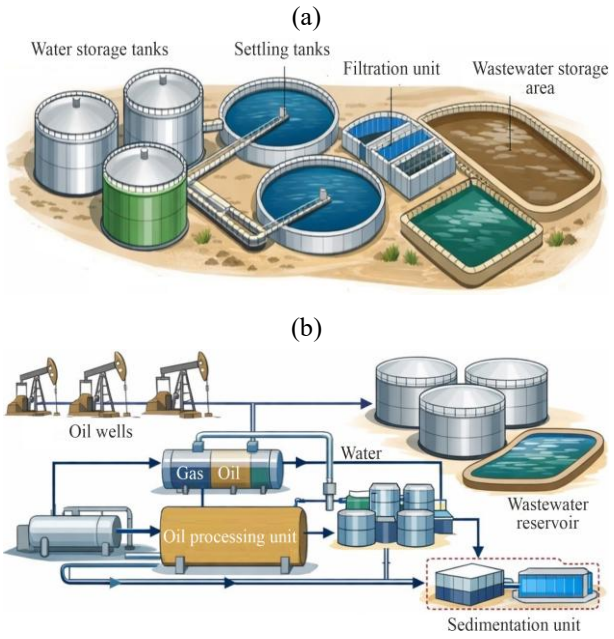


Figure 3. Water treatment and production infrastructure of the Kumkol oilfield: (a) water treatment facilities; (b) oil production and processing infrastructure

Wastewater generated at the Kumkol oilfield is primarily associated with oil-water separation processes, reservoir pressure maintenance systems, and equipment washing operations. This wastewater is characterized by elevated concentrations of dissolved salts, petroleum hydrocarbons, and trace elements in produced water streams. The total mineralization level ranged from 10 to 25 g/L, while petroleum hydrocarbon concentrations varied between 15 and 60 mg/L [64]. The concentration of suspended solids ranged from 20 to 120 mg/L, indicating the presence of mineral particles and corrosion products. The chemical composition of the wastewater and its environmental impacts are presented in Table 3.

The data presented in Table 3 show that the concentrations of petroleum hydrocarbons, total dissolved solids, chlorides, and several trace elements exceeded the permissible limits. High salinity and hydrocarbon content were identified as the main factors contributing to environmental impact.

Conventional treatment methods used at oil production facilities include gravity separation, flotation, filtration, and chemical coagulation. Gravity separation removed approximately 60-75% of free oil fractions, whereas flotation processes increased treatment efficiency to 80-90% [64], [65]. Filtration technologies enabled the removal of suspended solids at levels of 70-95%. However, complete removal of dissolved hydrocarbons and salts was not achieved.

Chemical treatment methods employing coagulants and demulsifying agents were used to destabilize oil-water emulsions and precipitate metal ions. The application of coagulants at doses of 30-100 mg/L enabled the removal of up to 75-92% of petroleum products. However, reagent consumption and sludge generation limited the overall efficiency of the process [66], [67].

Recent studies have shown that advanced oxidation processes, including plasma-based technologies, represent a promising approach for the treatment of complex oilfield wastewater [68]. During plasma treatment, the formation of reactive species promotes hydrocarbon oxidation, emulsion destabilization, and the transformation of dissolved contaminants. Therefore, the application of plasma technologies to multicomponent systems such as wastewater from the Kumkol oilfield constitutes an important research direction.

Overall, the analysis of wastewater composition and the treatment methods currently applied at the Kumkol oilfield revealed the limitations of conventional approaches and substantiated the need for advanced technologies capable of simultaneously removing high mineralization, hydrocarbon contamination, and trace elements. This further emphasizes the relevance of investigating plasma-based treatment technologies, which are addressed in the following sections of this study.

2. Methods

2.1. Research procedure

To evaluate the treatment efficiency of wastewater generated by mining and oil production using a high-frequency electrical discharge (plasma treatment), a conceptual scheme was employed that integrates contaminants, conventional treatment stages, and plasma-induced oxidation and transformation mechanisms (Fig. 4).

Table 3. Chemical composition of wastewater from the Kumkol oilfield and its environmental impacts

Component	Initial concentration, mg/L	Permissible level, mg/L	Environmental impact
Chlorides (Cl^-)	3500	350	Increased salinity of soils and groundwater
Sulfates (SO_4^{2-})	1200	500	Ecosystem disturbance and scale formation
Petroleum hydrocarbons	40	5	Toxic to aquatic organisms and reduce oxygen transfer
Iron (Fe^{2+}, Fe^{3+})	1.5	0.3	Contributes to corrosion and turbidity
Copper (Cu)	20	1.0	Toxic to aquatic organisms
Zinc (Zn)	15	5.0	Risk of bioaccumulation in living organisms
Calcium (Ca^{2+})	350	200	Increases hardness and scale formation
Magnesium (Mg^{2+})	180	100	Contributes to salinity and hardness
Ammonium (NH_4^+)	1.0	0.5	Indicates organic contamination
Total dissolved solids	15000	1000	Strong salinity impact on ecosystems
Suspended solids	80	25	Sedimentation and turbidity effects
Cadmium (Cd)	0.5	0.003	Highly toxic and bioaccumulative

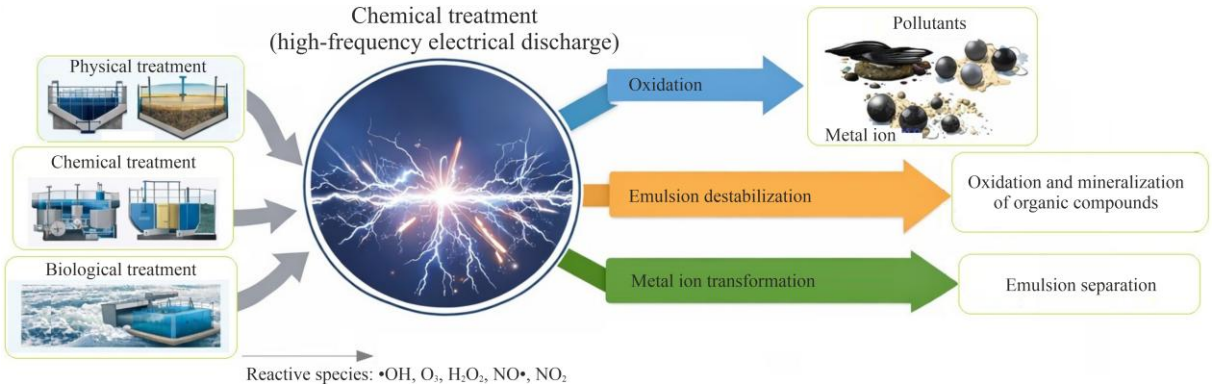


Figure 4. Schematic representation of the interaction between wastewater treatment methods and plasma oxidation mechanisms

The methodology presented in Figure 4 illustrates the relationship between physical, chemical, and biological treatment approaches and the role of reactive species generated by high-frequency electrical discharge (•OH, O_3, H_2O_2, and reactive nitrogen species) in oxidation, emulsion destabilization, and metal ion transformation processes. Wastewater samples collected from mining enterprises and the Kumkol oilfield were first characterized in terms of physicochemical properties and contaminant content. They were then subjected to plasma treatment in a laboratory-scale plasma-liquid reactor under controlled discharge conditions (10-25 kV, 10-30 kHz, interelectrode gap 3-7 mm, treatment time 5-30 min).

The dynamics of reactive species formation and contaminant removal efficiency were evaluated by comparing the initial and final water quality parameters. This made it possible to identify relationships among discharge conditions, reactive species generation, and treatment performance, and to formulate recommendations for industrial application.

2.2. Experimental setup

A laboratory-scale plasma-liquid reactor system was used to investigate the formation of reactive species (•OH, O_3, H_2O_2, and reactive nitrogen species) during the plasma treatment of wastewater from the mining and petroleum industries and to assess their role in contaminant transformation. A schematic diagram of the experimental setup is presented in Figure 5.

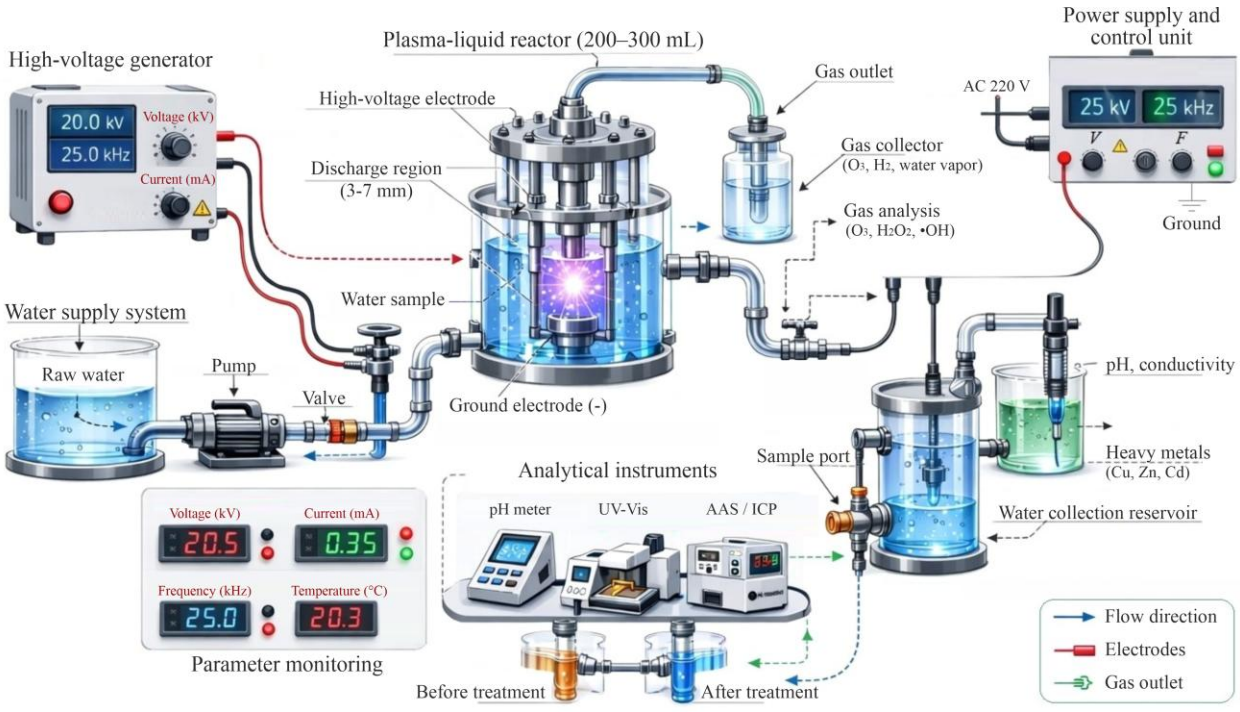


Figure 5. Schematic diagram of the experimental plasma-liquid water treatment system (10-25 kV, 10-30 kHz, interelectrode distance 3-7 mm, reactor volume 200-300 mL)

The experimental system consisted of the following main components:

– a water supply system including a raw water tank, a pump, and a control valve;

– a high-voltage power supply and control unit;

– a plasma-liquid reactor (200-300 mL)

– a gas outlet, collection, and analysis system;

– a treated water collection tank equipped with a sampling port;

– instruments for process monitoring and analytical measurements.

Wastewater samples collected from mining enterprises and the Kumkol oilfield were introduced into the reactor through a water supply system operating in either continuous

or semi-continuous mode. The plasma-liquid reactor was designed as a hermetically sealed chamber with a working volume of 200-300 mL. Inside the reactor, a high-voltage electrode (+) and a grounded electrode (–) were installed, with an adjustable interelectrode gap of 3-7 mm. This configuration ensured stable discharge generation and maintained the stability of the plasma-liquid interface.

Plasma treatment was generated using a high-frequency electrical discharge supplied by a high-voltage power source operating in the ranges of 10-25 kV and 10-30 kHz. Within the discharge zone, plasma-chemical reactions involving energetic electrons and excited species led to the dissociation of water molecules and the formation of reactive species. Published data and preliminary experimental results indicated hydroxyl radical (•OH) generation rates in the range of (1-5) 10^{-6} mol·L^{-1}·s^{-1}, ozone (O_3) concentrations from 10^{-6} to 10^{-4} mol·L^{-1}, and hydrogen peroxide (H_2O_2) accumulation from 10 to 80 mg/L. Owing to their oxidation potential of 2.07-2.80 V, these species created a strong oxidative environment that promoted the transformation and degradation of both organic and inorganic contaminants in multicomponent wastewater systems.

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During operation, the gas mixture generated in the reactor (O_3, H_2, and water vapor) was transported through the gas outlet line to a gas collector and subsequently analyzed using a gas analyzer to monitor reactive species formation. The treated water exited the reactor through the outlet channel into a water collection reservoir, where periodic sampling was carried out through a dedicated sampling port.

The process parameters, including voltage, current, frequency, and temperature, were recorded continuously using an integrated monitoring module. Water quality was assessed by measuring pH and electrical conductivity before and after treatment, while the concentrations of heavy metals (Cu, Zn, Cd) were determined using atomic absorption spectroscopy (AAS) and inductively coupled plasma (ICP) analysis. In addition, UV-Vis spectrophotometry was applied to monitor changes in the content of organic compounds.

Overall, the developed experimental setup made it possible to systematically investigate the relationships among discharge parameters (10-25 kV, 10-30 kHz, 3-7 mm), wastewater composition, reactive species generation intensity, and contaminant removal efficiency. The experimental data obtained were used to identify the optimal operating conditions that provide maximum treatment efficiency with minimized energy consumption.

2.3. Evaluation of treatment efficiency under laboratory conditions

To evaluate the efficiency of the plasma treatment process under laboratory conditions, the concentrations of heavy metals (Cu, Zn, and Cd) in wastewater from the mining and petroleum industries were determined before and after treatment. Plasma treatment was carried out in a plasma-liquid reactor with a high-frequency electrical discharge at a voltage of 10-25 kV, a frequency of 10-30 kHz, and an interelectrode gap of 3-7 mm. The treatment time ranged from 5 to 30 min depending on the experimental series.

The preliminary analysis showed that heavy metal concentrations in the wastewater exceeded the permissible limits. The concentration of copper (Cu) decreased from 20 to 0.5 mg/L after plasma treatment, corresponding to a removal efficiency of 97.5%. Similarly, the concentration of zinc (Zn) decreased from 15 to 0.4 mg/L, reaching a removal efficiency of 97.3%. The concentration of cadmium (Cd) was reduced from 0.5 to 0.02 mg/L, resulting in a treatment efficiency of 96%.

The obtained results demonstrated that the reactive species generated during plasma treatment (•OH, O_3, and H_2O_2), created a strong oxidative environment, which ensured the effective removal of heavy metal ions through oxidation, precipitation, and complexation mechanisms.

In addition to the experimental evaluation, a mathematical model was applied to describe the kinetics of heavy metal removal. Based on the laboratory data, it was assumed that the temporal decrease in metal ion concentration follows first-order reaction kinetics. This relationship can be expressed by the following Equation:

$$C(t) = C_0 \exp\left[-k_0 \Phi^a E^\beta t\right], \tag{14}$$

where:

$C(t)$ – contaminant concentration at time t, mg/L;
C_0 – initial concentration, mg/L;
K_0 – baseline kinetic constant;
Φ – flux of reactive species, or their effective concentration;
E – specific discharge energy or electric field intensity;
α, β – empirically determined influence coefficients;
t – treatment time, min.

The proposed model makes it possible to predict the dynamics of contaminant concentration changes during treatment and to determine the kinetic parameters on the basis of experimental data. According to the experimental results, the apparent reaction rate constant varied from 0.05 to 0.15 min^{-1} depending on the discharge parameters and water composition.

The treatment efficiency was calculated using the following Expression:

$$E(t) = \left(1 - \frac{C_t}{C_0}\right) \cdot 100 = \left(1 - \exp\left[-k_0 \Phi^a E^\beta t\right]\right) \cdot 100 . \tag{15}$$

This expression was used to quantify the removal performance for individual contaminants and to compare treatment efficiency under different discharge conditions.

3. Results

3.1. Analysis of physicochemical characteristics of wastewater from the mining and petroleum industries

To ensure a proper evaluation of plasma treatment efficiency, the initial physicochemical characteristics of wastewater collected from mining enterprises and the Kumkol oilfield were determined prior to the experiments. The obtained results showed that the investigated systems had a complex multicomponent composition, containing elevated

levels of dissolved salts, heavy metals, organic compounds, and suspended solids.

Wastewater from the mining industry was characterized by moderate mineralization, with total dissolved solids (TDS) ranging from 1.5 to 3.0 g/L. Heavy metal concentrations substantially exceeded the permissible limits, including copper (Cu) at 10-25 mg/L, zinc (Zn) at 8-18 mg/L, and cadmium (Cd) at 0.2-0.7 mg/L. The pH values varied from 6.2 to 7.5, indicating a slightly acidic to neutral medium. Electrical conductivity ranged from 2.1 to 4.8 mS/cm, confirming the presence of a high concentration of dissolved ions. Overall, Table 4 provides a more detailed description of the water quality parameters of mining wastewater prior to plasma treatment.

Table 4. Physicochemical characteristics of mining wastewater and comparison with permissible limits

No.	Parameter	Min	Max	Mean	Permissible limit	Environmental significance
1	Total dissolved solids (TDS), g/L	1.5	3.0	2.25	≤ 1.0	Increased salinity and ionic strength
2	Copper (Cu), mg/L	10	25	17.5	≤ 1.0	Toxic heavy metal with bioaccumulation potential
3	Zinc (Zn), mg/L	8	18	13.0	≤ 5.0	Trace element affecting aquatic ecosystems
4	Cadmium (Cd), mg/L	0.2	0.7	0.45	≤ 0.003	Highly toxic element
5	pH	6.2	7.5	6.85	6.5-8.5	Slightly acidic to neutral medium
6	Electrical conductivity, mS/cm	2.1	4.8	3.45	≤ 1.5	High concentration of dissolved ions

The obtained results indicate that mining wastewater contained total dissolved solids in the range of 1.5-3.0 g/L, copper concentrations of 10-25 mg/L, zinc concentrations of 8-18 mg/L, cadmium concentrations of 0.2-0.7 mg/L, and electrical conductivity values of 2.1-4.8 mS/cm, all of which exceeded the permissible levels. Although the pH remained within the range of 6.2-7.5, indicating a slightly acidic to neutral medium, the elevated concentrations of heavy metals and dissolved salts confirm a substantial degree of water contamination.

Wastewater from the Kumkol oilfield was identified as a more highly saline system compared with mining wastewater. Total dissolved solids ranged from 10 to 25 g/L, petroleum hydrocarbons from 15 to 60 mg/L, and suspended solids from 20 to 120 mg/L. Chloride concentrations were approximately 3500 mg/L, while sulfate concentrations reached 1200 mg/L. The pH values varied from 6.8 to 7.9, and electrical conductivity exceeded 15 mS/cm. Overall, Table 5 presents the initial physicochemical characteristics of wastewater from the Kumkol oilfield.

Table 5. Physicochemical characteristics of wastewater from the Kumkol oilfield and comparison with permissible limits

No.	Parameter	Min	Max	Mean	Permissible limit	Environmental significance
1	Total dissolved solids, g/L	10	25	17.5	≤ 1.0	High salinity and salt accumulation in soils and waters
2	Petroleum hydrocarbons, mg/L	15	60	37.5	≤ 5.0	Toxic organic contamination
3	Suspended solids, mg/L	20	120	70	≤ 25	Turbidity and sludge formation
4	Chlorides, mg/L	3500	3500	3500	≤ 350	Impact of high mineralization
5	Sulfates, mg/L	1200	1200	1200	≤ 500	Salinity and scale formation
6	pH	6.8	7.9	7.35	6.5-8.5	Neutral medium
7	Electrical conductivity, mS/cm	> 15	> 15	> 15	≤ 1.5	Very high ion concentration

The data presented in Table 5 indicate that wastewater from the Kumkol oilfield contained total dissolved solids in the range of 10-25 g/L, petroleum hydrocarbons at 15-60 mg/L, chlorides at 3500 mg/L, and sulfates at 1200 mg/L, all of which substantially exceeded the permissible limits. In addition, electrical conductivity above 15 mS/cm and suspended solids in the range of 20-120 mg/L confirm the high salinity and complex multicomponent contamination of this wastewater.

The obtained results indicate that both types of industrial wastewater exceeded the permissible limits for the major environmentally hazardous contaminants. Therefore, complete treatment using a single conventional method is problematic for such multicomponent systems with high ionic strength, which underscores the relevance and necessity of plasma-based technologies.

3.2. Formation of reactive species during plasma treatment

During plasma treatment using a high-frequency electrical discharge, the formation of highly oxidative reactive species in the liquid phase and at the plasma-liquid interface was confirmed experimentally. Plasma treatment in the laboratory-scale plasma-liquid reactor was carried out under operating conditions of 10-25 kV, 10-30 kHz, and 5-30 min. The main reactive species identified during treatment included hydroxyl radicals (•OH), ozone (O_3), hydrogen peroxide (H_2O_2), and reactive nitrogen species (RNS).

$$e^- + H_2O \rightarrow \bullet OH + H\bullet + e^-; \quad (16)$$

$$H_2O^+ + H_2O \rightarrow H_3O^+ + \bullet OH; \quad (17)$$

$$\bullet OH + \bullet OH \rightarrow H_2O_2; \quad (18)$$

$$H_2O_2 + h\nu \rightarrow 2\bullet OH; \quad (19)$$

$$e^- + O_2 \rightarrow 2O\bullet + e^-; \quad (20)$$

$$O\bullet + O_2 + M \rightarrow O_3 + M; \quad (21)$$

$$O_3 + H_2O \rightarrow \bullet OH + HO_2\bullet + O_2; \quad (22)$$

$$HO_2\bullet + HO_2\bullet \rightarrow H_2O_2 + O_2; \quad (23)$$

$$N_2 + O\bullet \rightarrow NO + N\bullet; \quad (24)$$

$$NO + O\bullet \rightarrow NO_2. \quad (25)$$

A pronounced time-dependent increase in H_2O_2 concentration was observed, with its level rising progressively during treatment and reaching 10-80 mg/L (Fig. 6).

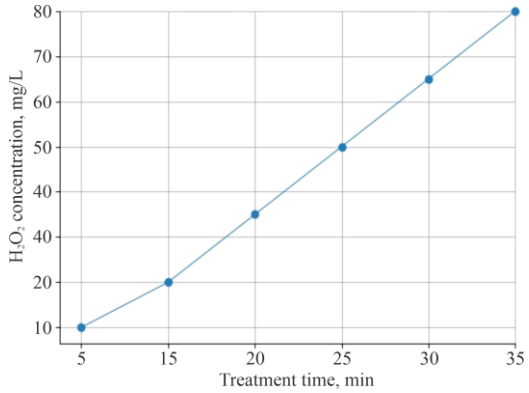


Figure 6. Temporal evolution of H_2O_2 concentration during plasma treatment

Ozone (O_3) was generated in the gas phase, and part of it dissolved into the liquid phase, where it participated in the oxidation of organic contaminants. The intensity of hydroxyl radical (•OH) formation was estimated using indirect methods, indicating a generation rate on the order of (1-5) 10^{-6} mol·L^{-1}·s^{-1}.

The graph shows a gradual increase in H_2O_2 concentration with increasing plasma treatment time, from approximately 10 mg/L after 5 min to 80 mg/L after 30 min. This trend indicates the continuous formation of reactive species at the plasma-liquid interface and the progressive intensification of the oxidative environment with increasing treatment duration.

The results demonstrated that plasma treatment of multicomponent industrial wastewater generates a strong oxidative environment, and that the primary treatment effect is governed by the formation of these reactive species.

3.3. Relationship between discharge parameters and the intensity of reactive species formation

The experimental results showed that the intensity of reactive species formation strongly depended on the discharge parameters. An increase in discharge voltage led to enhanced generation of •OH and O_3, whereas operation within the selected frequency range ensured stable discharge conditions and promoted the accumulation of reactive species. The selected interelectrode distance was found to be suitable for maintaining stable plasma generation, thereby ensuring efficient production of hydroxyl radicals and hydrogen peroxide. In addition, saturation of reactive species concentration was observed under high-energy operating conditions. Overall, Figure 7 illustrates the effect of discharge voltage on the generation of reactive oxygen species.

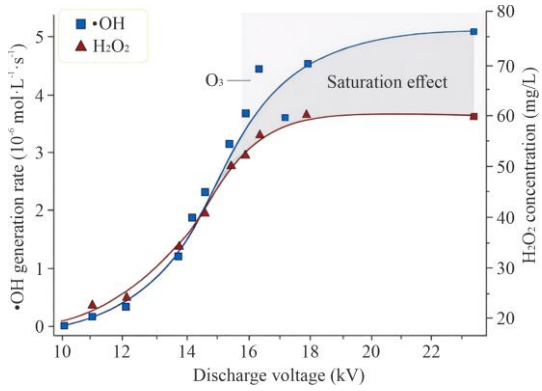


Figure 7. Effect of discharge voltage on the generation of reactive oxygen species

The results show that at a discharge voltage of 10 kV, the •OH generation rate was approximately 1 10^{-6} mol·L^{-1}·s^{-1}, while the H_2O_2 concentration was about 10 mg/L. When the voltage was increased to 20 kV, the •OH generation rate rose to 3-4·10^{-6} mol·L^{-1}·s^{-1}, and the H_2O_2 concentration increased to 45-60 mg/L. At 25 kV, the •OH generation rate reached approximately 5 10^{-6} mol·L^{-1}·s^{-1}, while the H_2O_2 concentration increased to 70-80 mg/L. Beyond this point, a saturation effect was observed due to radical recombination and decomposition processes, which led to a reduced growth rate.

3.4. Influence of reactive species on contaminant removal efficiency

The results of the study showed that contaminant removal efficiency during plasma treatment was directly related to the intensity of reactive species formation (•OH, O_3, and H_2O_2). Treatment in the plasma reactor led to a significant decrease in heavy metal concentrations (Table 6). Laboratory experiments showed that the concentration of copper ions (Cu) decreased from an initial value of 20 to 0.5 mg/L after treatment, corresponding to a removal efficiency of 97.5%. Similarly, the concentration of zinc (Zn) decreased from 15 to 0.4 mg/L, resulting in a removal efficiency of 97.3%, while the concentration of cadmium (Cd) decreased from 0.5 to 0.02 mg/L, corresponding to a removal degree of 96%.

Table 6. Heavy metal removal efficiency during plasma treatment

Contaminant	Cu	Zn	Cd
Initial concentration C_0, mg/L	20	15	0.5
Final concentration, C, mg/L	0.5	0.4	0.02
Removed amount, ΔC, mg/L	19.5	14.6	0.48
Residual fraction C/C_0, %	2.5	2.7	4
Removal efficiency E, %	97.5	97.3	96

The results showed that the reactive species generated during plasma treatment effectively removed heavy metal ions through oxidation, precipitation, and complexation processes, while organic contaminants were degraded by 70-90%. Kinetic analysis indicated that contaminant removal followed a first-order model and increased with discharge energy, confirming plasma treatment as a promising technology for the simultaneous removal of heavy metals and organic contaminants from multicomponent aqueous systems.

4. Discussion

The experimental results obtained in this study confirmed that plasma treatment based on a high-frequency electrical discharge creates an intense oxidative environment in multicomponent wastewater, which determines the observed contaminant removal efficiency. As shown in Figure 6, the progressive increase in hydrogen peroxide concentration during treatment indicates the continuous formation and accumulation of reactive species at the plasma-liquid interface. This behavior can be explained by the sequence of electron-induced water dissociation reactions (Equations (16)-(23)), which lead to hydroxyl radical formation followed by recombination processes.

The generation of •OH radicals at rates of (1-5) 10^{-6} mol·L^{-1}·s^{-1} and the accumulation of H_2O_2 up to 80 mg/L provided strong oxidative conditions that promoted the degradation of organic compounds and the transformation of dissolved heavy metals.

The relationship between discharge parameters and the intensity of reactive species formation, as observed in Figure 7, further explains the treatment efficiency obtained. Increasing the discharge voltage raised electron energy and collision frequency, thereby intensifying water dissociation reactions and oxygen activation. However, the saturation effect observed at higher voltages can be attributed to radical recombination processes and energy dissipation mechanisms, which limit any further increase in reactive species concentration.

The kinetic model expressed by Equation (14) successfully described the temporal evolution of contaminant concentration, whereas Equation (15) quantitatively linked treatment efficiency to discharge energy and reactive species flux. The agreement between the experimental removal efficiencies presented in Table 6 and the model predictions confirmed the adequacy of the proposed kinetic approach for describing plasma treatment processes in multicomponent systems.

Comparison of the obtained results with existing wastewater treatment technologies revealed several distinctive features of the proposed method. As shown in the comparative analysis of conventional methods (Table 1), physical processes mainly remove suspended solids, whereas chemical and biological methods demonstrate limited effectiveness in highly mineralized media. In contrast, plasma treatment achieved removal efficiencies exceeding 95% for heavy metals (Table 6) and up to 85-98% degradation of organic compounds, which is consistent with the literature on advanced oxidation processes. Moreover, unlike coagulation-based technologies, the plasma method does not require chemical reagents and generates substantially less sludge, thereby improving environmental sustainability. The specific energy consumption values of 0.4-0.9 kWh/m^3 presented in Table 2 are comparable to, or lower than, those reported for ozonation and UV/H_2O_2 processes in previous studies, highlighting the competitive energy efficiency of the electrical discharge method.

Despite these promising results, several limitations of the present study should be noted. First, the experiments were conducted at laboratory scale with reactor volumes of 200-300 mL, which limits the direct extrapolation of the results to industrial systems. Second, the investigated wastewater samples represented specific conditions of the mining and petroleum industries, implying that variations in ionic composition, buffering capacity, and contaminant load may affect the kinetics of reactive species formation. Third, indirect methods were used to estimate hydroxyl radical formation, which may introduce uncertainty into the quantitative interpretation of the results. In addition, the applicability of the kinetic model is limited to the investigated range of discharge parameters (10-25 kV, 10-30 kHz, 3-7 mm), and further validation is required outside this operating window.

In addition to these limitations, several shortcomings of the study should also be acknowledged. The experimental setup did not provide real-time monitoring of all reactive species, which limits the mechanistic interpretation of plasma-liquid interactions. In addition, no gas-phase plasma diagnostics were performed, restricting understanding of the relationship between gas-phase and liquid-phase chemistry. These shortcomings can be addressed in future studies through the use of optical emission spectroscopy, radical probing techniques, and advanced analytical methods that allow direct measurement of transient species.

The present study represents an important step toward the development of plasma-based wastewater treatment technologies; however, further research is required for their practical implementation. Future work may include reactor scale-up, optimization of electrode configurations and operating parameters, and integration of plasma processes with conventional treatment stages to create hybrid systems. Mathematical challenges related to the modeling of multicomponent plasma-liquid systems should also be addressed, including coupled transport-reaction processes and the stochastic behavior of the discharge. Experimental difficulties may arise in maintaining discharge stability in highly conductive media and in controlling energy efficiency during scale-up. Nevertheless, overcoming these challenges will contribute to the development of environmentally safe and energy-efficient plasma technologies for the treatment of complex industrial wastewater.

5. Conclusions

This study investigated the regularities of reactive species formation during the plasma treatment of multicomponent wastewater generated by mining and oil production activities and evaluated their role in contaminant removal. The results showed that the initial composition of the studied wastewater was characterized by substantial mineralization (1.5-25 g/L) and elevated concentrations of heavy metals (Cu, Zn, Cd), petroleum hydrocarbons, and dissolved salts exceeding the permissible limits. These characteristics confirmed the limitations of conventional treatment methods and substantiated the need for advanced oxidation technologies.

The experimental investigations confirmed that plasma treatment using a high-frequency electrical discharge led to the formation of hydroxyl radicals (•OH), ozone (O_3), hydrogen peroxide (H_2O_2), and reactive nitrogen species at the plasma-liquid interface. The •OH generation rate was estimated to be within $(1\text{-}5)\cdot10^{-6}$ mol·L^{-1}·s^{-1}, ozone concentration ranged from 10^{6} to 10^{-4} mol·L^{-1}, and hydrogen peroxide accumulation reached 10^{-8} mg/L. The formation of these reactive species created a strong oxidative environment that promoted the effective transformation and degradation of both organic and inorganic contaminants in wastewater.

The results demonstrated a clear dependence of reactive species formation on discharge parameters. Stable plasma generation and efficient production of reactive species were achieved within the operating range of 10-25 kV, 10-30 kHz, and an interelectrode gap of 3-7 mm. Increasing the discharge voltage enhanced reactive species formation; however, a saturation effect was observed at higher energy levels because of radical recombination processes, indicating the importance of discharge parameter optimization for maximizing treatment efficiency.

The evaluation of contaminant removal efficiency confirmed the high performance of plasma treatment. Copper concentration decreased from 20 to 0.5 mg/L (97.5%), zinc from 15 to 0.4 mg/L (97.3%), and cadmium from 0.5 to 0.02 mg/L (96%). In addition, the degradation of organic compounds reached 70-90%. The applied kinetic model successfully described the exponential decrease in contaminant concentration over time and made it possible to quantitatively assess the influence of reactive species flux and discharge energy on treatment efficiency.

A comparative analysis with conventional physical, chemical, and biological treatment methods revealed several

advantages of plasma technology, including the absence of chemical reagents, reduced sludge formation, simultaneous removal of multicomponent contaminants, and competitive energy consumption in the range of 0.4-0.9 kWh/m^3. These characteristics demonstrate the environmental sustainability and technological feasibility of plasma treatment for complex industrial wastewater.

Overall, the conducted study provided insight into the mechanisms of reactive species formation during plasma treatment, established relationships between discharge parameters and treatment efficiency, and confirmed the potential of plasma technology for advanced wastewater treatment in the mining and petroleum industries. Future research should focus on reactor scale-up, optimization of electrode configurations and operating parameters, development of hybrid treatment systems, and validation of plasma technology under industrial operating conditions.

Author contributions

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

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Conflicts of interest

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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Формування реактивних видів під час плазмової обробки стічних вод гірничодобувної та нафтогазової промисловості

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Мета. Вивчення закономірностей формування реактивних видів під час плазмової обробки багатокомпонентних стічних вод гірничодобувної та нафтогазової промисловості, а також оцінка їх ролі в трансформації забруднювачів та ефективності їх видалення.

Методика. Лабораторні експерименти проводилися із використанням спеціально розробленого плазмово-рідинного реактора, що працював в умовах високочастотного електричного розряду (10-25 кВ, 10-30 кГц, міжелектродний зазор 3-7 мм). У ході дослідження визначали вихідні фізико-хімічні характеристики стічних вод, зокрема pH, електричну провідність, загальний вміст розчинених речовин і концентрації важких металів (Cu, Zn, Cd). У процесі плазмової обробки аналізувалися утворення реактивних видів (•OH, O_3, H_2O_2), а для опису динаміки видалення забруднювачів та ефективності очищення було застосовано кінетичну модель.

Результати. Встановлено, що плазмова обробка призводить до утворення гідроксильних радикалів зі швидкістю (1-5)·10^{-6} моль·$л^{-1}$·$с^{-1}$, озону в концентраційному діапазоні 10^{-6}-10^{-4} моль·$л^{-1}$ та накопичення пероксиду водню в межах 10-80 мг/л, що формує виражене окиснювальне середовище. Концентрація міді знизилася з 20 до 0.5 мг/л (97.5%), цинку з 15 до 0.4 мг/л (97.3%), кадмію з 0.5 до 0.02 мг/л (96.0%). Ступінь розкладання органічних забруднювачів становив 70-90%. Показано, що інтенсивність формування реактивних видів істотно залежить від параметрів розряду, а запропонована кінетична модель адекватно описує експериментально спостережувану динаміку очищення.

Наукова новизна. Дослідження являє собою комплексний експериментально-модельний аналіз формування реактивних видів під час плазмової обробки високомінералізованих багатокомпонентних стічних вод гірничодобувної та нафтогазової промисловості. Встановлено кількісні залежності між параметрами розряду, генерацією реактивних видів та ефективністю видалення забруднювачів.

Практична значимість. Отримані результати підтверджують перспективність плазмових технологій для глибокого очищення промислових стічних вод складного складу та високої солоності. Практична реалізація запропонованого підходу може сприяти підвищенню екологічної безпеки, зниженню витрат реагентів, зменшенню утворення осадів і розширенню можливостей повторного використання води у гірничодобувних і нафтовидобувних процесах.

Ключові слова: *плазмова обробка; стічні води; електричний розряд; реактивні види; важкі метали; стічні води гірничодобувної промисловості; нафтопромислові стічні води*

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