

INVESTIGATION OF CHARACTERISTICS OF A BARRIER DISCHARGE IN A WATER-AIR ENVIRONMENT

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Abstract- In this article, studies of the emission spectra of a pulsed barrier discharge in a water-air medium are considered. The main short-lived active particles formed in the plasma discharge were determined and its main characteristics were evaluated. The purpose of this work is to identify the main short-lived particles formed during the electric discharge treatment of water, as well as to estimate the discharge parameters by the method of emission spectroscopy. The results obtained can be directly used to elucidate the processes occurring in an electric discharge, and also as initial data in mathematical modeling of the physical pattern and chemical transformations in a barrier discharge.

Keywords: Water-Air Environment, Emission Spectrum, Spectral Region, Emission Bands, Micro Discharge.

1. INTRODUCTION

The high degree of pollution of surface and ground waters and the growing demands on water quality cause a significant need for water treatment systems that would be efficient, easy to operate and would not eventually lead to environmental pollution due to the use of toxic chemicals.

Among the environmentally friendly water purification technologies, ozonation is widely used [1]. Ozone is a strong oxidizing agent that is industrially produced from air or oxygen by means of an electrical discharge. Ozonation has a number of disadvantages, the main of which is the inefficient use of energy. The synthesis of ozone in an electric discharge is a reversible reaction, while a significant part of the energy is lost to maintain the balance between ozone and oxygen. An increase in the efficiency of water purification compared to traditional ozonation is possible with the combined use of ozone and other active particles, as well as ultraviolet radiation generated in the discharge. From this standpoint, the discharge in moist air and water-air environment is of interest. In this case, the products of electric discharge treatment are ozone, hydrogen peroxide, and various kinds of radicals: OH, O(³P), O(¹D), O₂(b¹Σ_g⁺), O₂⁺, O⁻, nitrogen-containing, and some other particles [2-5]. Under the influence of active particles, most of the impurities contained in the water are oxidized. Organic substances are oxidized up to complete mineralization (CO₂ and H₂O), or to the formation of less toxic products.

Studies by a number of authors [2-4] point to the possibility of increasing the efficiency of processing when switching to pulsed discharges. Thus, it was shown in [2] that a barrier discharge powered by nanosecond pulses has advantages in the purification of moist air from a number of organic impurities and ammonia, which is associated with a higher uniformity of this discharge. At the same time, it is noted that the active radicals formed during the processing have many times greater reactivity than ozone and ultraviolet radiation.

In [3], the effect of a pulsed corona discharge on aqueous solutions of phenol was considered. It is shown that in the case of a corona discharge above the water surface, the efficiency of electric discharge treatment is close to the efficiency of water ozonation. At the same time, the generated amount of ozone is significantly less than in the ozonation technology. The authors explain the high purification efficiency by the action of short-lived active particles, in particular, OH hydroxyl radicals. At the same time, it should be noted the relatively high complexity of installations based on a pulsed corona discharge and, accordingly, limited practical applicability.

One of the most promising and practically easy to implement electric discharge methods of water purification is the use of a pulsed barrier discharge in a water-air flow [4]. The installation, in which the principle of barrier discharge is implemented, has a high efficiency in water purification from organic impurities (in particular, phenol and trichloroethylene), as well as iron and manganese compounds, which are often found in groundwater. At the same time, the flow mechanism and the main acting factors of the discharge in a water-air medium have not been sufficiently studied due to the high degree of non-equilibrium of the discharge plasma, as well as high rate of elementary processes in discharge.

2. EXPERIMENTAL METHODOLOGY

It is known that a barrier discharge is a discharge in a gas that occurs under the influence of a voltage applied to the electrodes, if at least one of the electrodes is covered with a dielectric [5]. In such a discharge, charge transfer occurs in separate channels, called micro discharges, which are distributed over the entire volume of the interelectrode gap.

The discrete structure of the barrier discharge and the short lifetime of the discharge products make it difficult to study the main processes in the discharge plasma. One of the main research methods in this case is the spectral method. The study of the spectra of the barrier discharge is complicated by the low intensity of the glow, as well as the significant non-stationarity of the discharge when water drops are introduced into the discharge gap.

Therefore, a much smaller number of works are devoted to the study of radiation in this type of discharge than, for example, to spectral studies of a glow discharge or arc. The measurements were carried out by the method of emission spectroscopy. The method of emission spectroscopy can provide not only reliable information on the presence of particles, for which the discharge energy is sufficient, but also allows one to estimate a number of the most important plasma parameters, such as the average electron energy, plasma temperature, and electric field strength.

2.1. Installation for the Study of Emission Spectra

The scheme of the discharge chamber used in the experiments is shown in Figure 1a. Water was sprayed into drops about 3 mm in diameter and fed to the electrode system. The discharge was created between wire electrodes 2 mm in diameter enclosed in quartz glass tubes with a wall thickness of 1.5 mm (Figure 1b). The width of the air gap between them was 3 mm. Air was pumped through the electrode system in the direction opposite to the water flow at a rate of 5-10 l/min. A quartz window was used to observe discharge radiation.

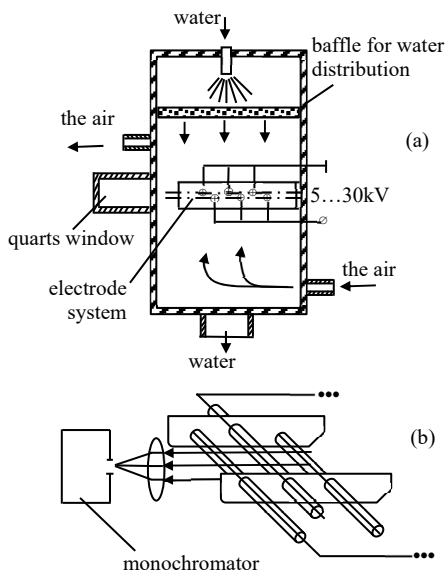


Figure 1. (a) Discharge chamber for observing the spectral characteristics of the discharge, and (b) the design of the electrode system used in the experiments

The emission spectra were recorded using an MDR-2 monochromator (Figure 2a). To record the spectra, photo-electronic multiplier tubes (PEMs) were used, the sensitivity range of which lies in the band from 200 to 650

nm and from 400 to 800 nm. Before the experiments, the PEM was calibrated using reference radiation sources. The PMT signal was amplified by a V6-9 selective amplifier and fed to the input of a storage oscilloscope (C9-8, Tektronix TDS-220). Thus, the setup made it possible to study the radiation spectrum of the barrier discharge in the wave lengths range (200-800 nm).

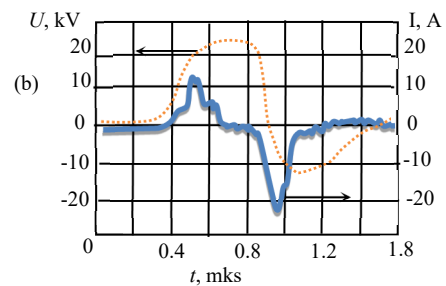
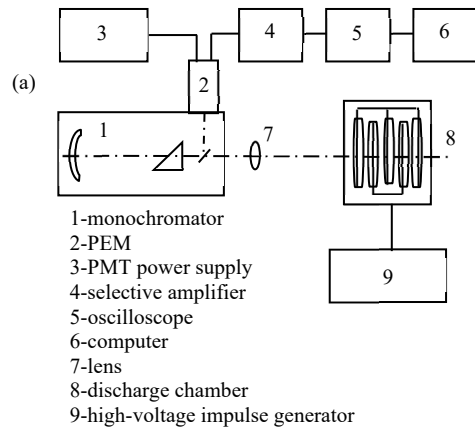


Figure 2. (a) Installation scheme for observing the spectral characteristics of the discharge, and (b) oscillograms of the current and voltage in the discharge

To power the electrode system, we used a nanosecond pulse generator consisting of a low-voltage thyristor unit and high-voltage magnetic compression links that form the output pulse [6]. The output capacitance of the generator was 150 pF. A pulsed voltage with an amplitude of 15-30 kV, a pulse duration of 500..600 ns, and a repetition rate of 1 kHz was applied to the electrodes. In the experiments, the electrical parameters of the installation were controlled - current, voltage, and output power. Measurements were made using a resistive divider, a low inductance current shunt, and a Tektronix TDS220 oscilloscope with up to 100 MHz bandwidth. Figure 2b shows the *I* and *U* waveforms on the electrode system.

The oscillograms were used to determine the barrier discharge ignition voltage, which was about 16.5 kV for the electrode system used in the experiments. At this voltage, the appearance of appreciable ozone concentrations at the output of the setup was recorded, and the glow of the discharge gap also arose. When the discharge ignition voltage was reached, the current in the circuit increased sharply. The energy input into the discharge gap at a voltage $U=25$ kV, calculated by integrating current and voltage oscillograms, was 45 mJ/pulse.

3. RESULTS

The barrier discharge in dry air and in air saturated with water vapor consisted of many micro discharges. The introduction of water drops led to a noticeable change in the electrical characteristics of the system: a decrease in the discharge ignition voltage, as well as an increase in the discharge current within 20%. The emission spectrum of the barrier discharge plasma was studied in wave lengths range (200-800 nm). Figure 3 shows discharge spectrum in a water-air environment at atmospheric pressure.

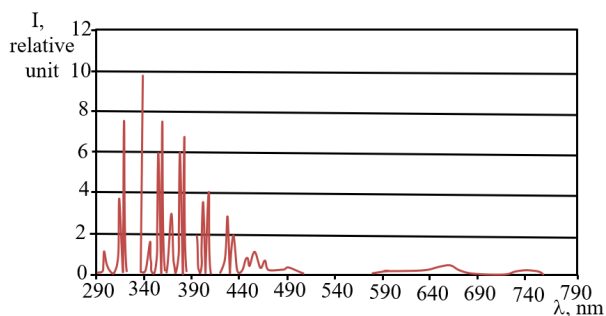


Figure 3. Emission spectrum of air/water system in a barrier discharge

It can be seen from the Figure that the largest number of emission bands of different intensities lies in the wavelength region from 290 to 460 nm. In the region of the spectrum where the wave length is 500...800 nm, broad low-intensity emission bands are recorded.

- Spectral region 280-320 nm; In this region of the spectrum, three broad emission bands are recorded with maxima at 297.5 nm, 313.75 nm, and 316.0 nm (Figures 3 and 4). Mathematical processing of the spectra revealed the presence of a fine structure of the emission bands. So, in the wavelength range of 292-299 nm, 4 bands are visible with maxima at 292.5, 293.5, 296.25 and 297.5 nm (Figure 4), and in the wavelength range of 310...316 nm, 5 emission bands - 310.8, 312.3, 313.4, 315.5, 316 nm. Of these bands, only the 313.7 and 316.0 nm lines are narrow.

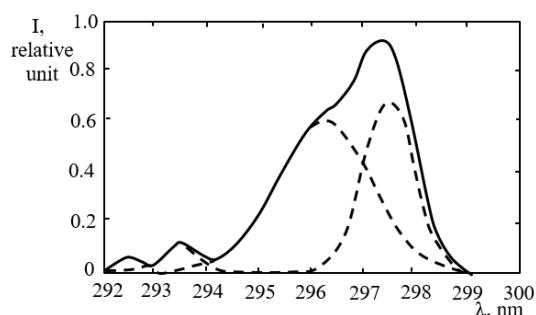
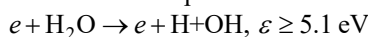


Figure 4. Emission spectrum of OH-radicals. Experimental observations (—) and results of mathematical processing (- -)

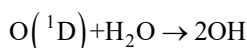
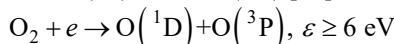
According to the literature data [7], the studied regions contain emission lines of OH radicals, as well as electronic-vibrational transitions of the molecular nitrogen of the 2⁺ system. The broad emission bands are responsible for the electronic-vibrational transitions OH (A-X; 0-0). 310.8 nm OH (A-X; 0-0); 312.3 nm OH (A-X; 0-0); 315.5 nm OH (A-X; 1-1) and 296.25 nm OH (A-X; 1-0), 294.00 nm OH (A-X; 2-0), 292.5 nm OH (A-X; 2-0) [13]. The bands we studied, which are responsible for the hydroxyl

radical (OH), are significantly shifted to the region of spectral long waves. The authors of [8], in a He/H₂O mixture in the study of glow discharge plasma, found a similar shift of OH-radical emission lines to the long-wavelength region and explained this by the presence of cluster molecules (OH)_n^{*}, which are formed when two or more OH radicals collide. One of the OH radicals is in an excited state. The presence of hydroxyl radicals of the barrier discharge is also confirmed by a weak band in the region of 660-670 nm.

The intensity of these emission bands increases as the concentration of water vapor in the air increases. At a humidity of 60%, saturation occurs, which is consistent with the results obtained in [9]. At the same time, intensity of the band is directly proportional to the current density and increases without saturation. This behavior indirectly indicates that formation of OH radicals can occur due to direct electronic processes of water excitation [10]:



and/or by transferring the excitation of the excited oxygen atom O(¹D) to water (2.3) [10]:



- The wavelength range is 320-600 nm; As can be seen from Figure 3, in this region of the spectrum, a number of intense emission lines are observed, which belong to the electronic-vibrational transitions N₂. In the emission spectra of the discharge, the band of molecular nitrogen 337.1 nm of the 2⁺ system of nitrogen is well identified: the transition N₂(C³Π_u, v) → N₂(B³Π_g, v), as well as the bands of the 2⁺ system of nitrogen N₂ (λ=380.75 nm and 400.25 nm).

When the voltage on the electrodes is increased to 23 kV, a weak nitrogen ion line appears (1⁻ nitrogen system: transition N₂⁺(B²Σ_u⁺, v) → N₂⁺(X²Σ_g⁺, v) λ = 391.4 nm,

but its intensity is approximately 100 times less than the linear 337.1 nm. At voltages on the electrodes above 26 kV, weak emission bands appear in the wavelength region of 236.3 and 237.0 nm (Figure 5), which are related to electronically excited NO molecules (transition 0→1 of the γ- system of nitric oxide). In the spectral region 425-490 nm, several lines of low intensity are observed. It is not possible to identify all the lines, however, we believe that the weak bands at 434.0 nm and 486.1 nm are responsible for the emission of hydrogen atoms of series H_γ and H_β, respectively.

The concentration of ozone in the water-air environment is several times lower than the concentration of ozone in the air. Under the experimental conditions, the energy consumption for the production of ozone in air with a humidity of 25% was 10-14 kWh/kg at an ozone concentration of 5-9 mg/l. Upon transition to a discharge in a water-air flow, the ozone concentration in the air at the outlet of the reactor noticeably decreased. Energy consumption for ozone production amounted to 40-70 kWh/kg.

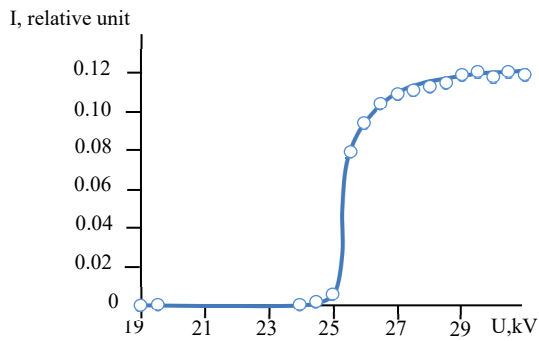


Figure 5. Dependence of the intensity of NO radiation $\lambda=236.3$ nm on the applied voltage

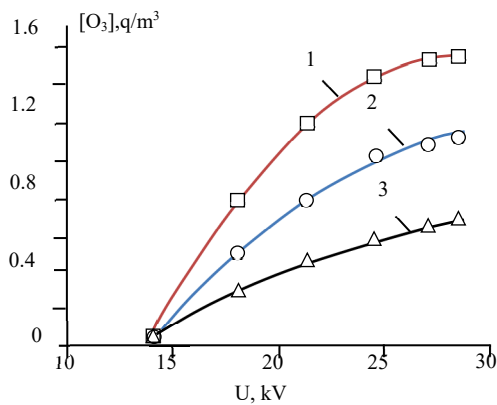


Figure 6. Dependence of the ozone concentration on the applied voltage, Air flow rate: 1) 0.3, 2) 0.62, 3) 1.2 m³/h

With an increase in the voltage amplitude, the dependence of the ozone concentration on the voltage experienced saturation at an ozone concentration of about 1.5 mg/l (Figure 6). The ozone concentration in the discharge-treated water was 0.1-0.2 mg/l.

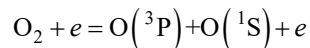
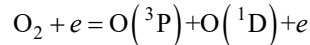
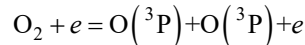
4. DISCUSSION OF RESULTS

Excitation of nitrogen. Of particular interest are the emission bands $N_2(C^3\Pi_u \rightarrow B^3\Pi_g)$ and $N_2^+(B^2\Sigma_u^+ \rightarrow X^2\Sigma_g^+)$. From the emission spectrum of the barrier discharge (Figure 3) that these emission lines do not overlap with each other, and the electron energies required to generate particles $N_2(C^3\Pi_u)$ and $N_2^+(B^2\Sigma_u^+)$ are different and amount to 11.0 and 18.7 eV, respectively. Due to the large difference in the energies of the electrons required for the formation of these particles, the ratio of intensity $I(N_2^+)/I(N_2)$ depends significantly on the average electron energy in the discharge and electric field intensity and can be used to calculate these parameters [11, 12].

Using the dependences proposed by the authors [11], we estimated the electric field strength and the average electron energy, which amounted to about 100 kV/sm and 6 eV, respectively, which is an order of magnitude higher than the breakdown voltages during a discharge in air at atmospheric pressure.

In our case, when a voltage with an amplitude of 20 kV or less was applied to the electrodes, the emission band at 391.4 nm was not recorded. Considering that an energy of 18.7 eV is required to excite $N_2^+(B^2\Sigma_u^+)$, it can be assumed that there are no electrons with such an energy in the plasma. This allows you to determine the requirements for the supply voltage to reduce the production of nitrogen oxides in water treatment process.

Estimation of temperature of gas molecules. The average gas temperature in the discharge chamber, measured with a thermometer, was 300 ± 1 K, which is 2-3 degrees higher than the air temperature at the installation inlet. Measuring the local gas temperature in the discharge gap is a more serious problem. The exact value of the temperature of heavy particles in the discharge can be determined from the rotational temperature of the state $N_2(C^3\Pi_{u,v=0})$ with a wavelength $\lambda=337.1$ nm [5]. The small width of this line indirectly indicates a low plasma temperature, which practically does not differ much from 300K. Estimates have shown that the plasma temperature does not exceed 350 K. This value, apparently, corresponds to the plasma temperature inside the micro discharge. Determination of dissociation constants of possible oxygen decomposition reactions. Using the obtained values of the E_{max} in the discharge gap $E \approx 100$ kV/sm, the rate constants of molecular oxygen dissociation were determined by expressions [10]:



$$lgk = -7.9 - 13.4 / E$$

$$lgk = -8.0 - 16.9 / E$$

$$lgk = -8.8 - 11.9 / E$$

$$k = 1.13 \times 10^{-8} \text{ cm}^3 \text{ s}^{-1}$$

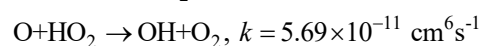
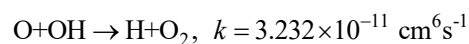
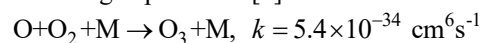
$$k = 8.78 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$$

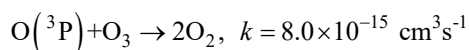
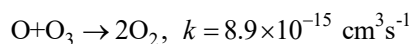
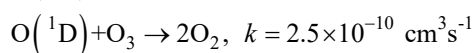
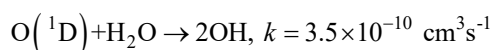
$$k = 1.45 \times 10^{-9} \text{ cm}^3 \text{ s}^{-1}$$

The results obtained are in good agreement with the corresponding reaction constants given in the literature [5]. Using k , one can determine the oxygen dissociation rate [10]:

$$v_{dis} = [O_2] n_e k = 6 \times 10^{18} \times 2 \times 10^7 \times 1.13 \times 10^{-8} = 1.32 \times 10^{18} \text{ cm}^{-3} \text{ s}^{-1}$$

In this case, the lifetime of excited oxygen atoms lies within 10^{-4} s. The resulting atomic oxygen interacts with impurity particles and molecules of the surrounding gas. According to [1], the main ways of spending atomic oxygen in the absence of impurities are determined by the following expressions [1]:





Theoretically, an increase in the absorption of active particles in the discharge zone is possible by creating turbulence and a developed contact surface in the water-air flow. The lifetime of OH-radicals in an ozone-air mixture, determined experimentally, is 0.1-0.3 ms, the diffusion radius of OH-radicals is about the diameter of a microdischarge [14]. At a maximum linear air flow velocity of 0.1 m/s, OH-radicals during their lifetime are transferred to a distance of no more than 30 μm , while their concentration decreases by more than 10 times. Therefore, air movement cannot make a significant contribution to the processes of mass transfer of OH radicals. To be transferred to water, radicals must be created in the immediate vicinity of the water surface.

Experimental observations show that the discharge is ignited near water droplets located in the interelectrode gap. This is also indicated by the results of numerical simulation of the electric field in a water-air medium performed in. Due to the high relative permittivity of water, the electric field strength increases in the gas volume surrounding the drop. The discharge occurs in areas with the highest field strength, while the discharge channel is in contact with the surface of a water drop.

When the interelectrode gap is closed by a water drop, almost all the voltage is applied to the dielectric barriers and the field strength on the drop surface decreases to values less than 1.5 kV/cm, which is below the discharge ignition threshold.

The results obtained when considering the mechanism of chemical processes, taking into account the above reactions, can later be used in modeling the processes of water purification in a barrier discharge plasma. An adequate description of the mechanism of water purification requires knowledge of the main characteristics of the discharge, as well as taking into account all the main reactions occurring in the discharge gap. Obviously, spectral studies and theoretical modeling should be confirmed by chemical studies of the initial and final products of electric discharge processing. Therefore, they should be considered in combination with other methods for studying the processes occurring during water purification in a pulsed discharge.

5. CONCLUSION

The electrical parameters of a pulsed barrier discharge in a water-air flow differ from the characteristics of a discharge in air. In a water-air environment, an increase in active power and a decrease in the discharge ignition voltage are observed [15].

1) Investigations of the emission spectrum of a pulsed barrier discharge in an air-water medium were carried out. It is shown that the main primary products of the action of a nanosecond pulsed barrier discharge on an air-water

mixture, according to emission spectroscopy data, are OH radicals and excited oxygen atoms.

2) Based on the emission spectra of a barrier discharge in an air-water medium the electric field strength and the average energy of accelerated electrons are estimated, which are 100 kV/cm and 6 eV, as well as the temperature of molecules and ions in the discharge, which turned out to be close to 300 K.

3) The average energy of electrons in the discharge under study is below the threshold energy necessary for excitation of elementary reactions of formation of nitrogen oxides. Therefore, when treating water in a pulsed barrier discharge at voltages below 26 kV, nitrogen-containing products are absent.

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