

# Electric Discharges and the Prolonged Microbial Resistance of Water

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**Abstract**—One of alternative methods of water disinfection is its treatment by pulsed electric discharges (PEDs). In this case, during the discharges, bacteria are destroyed by UV radiation with a significant share of energy in waves of 200–400 nm and by shock waves, which are formed at the initial stage of the discharge-column widening. This paper describes the electrophysical properties of PED in water, presents the results of an investigation of the physical, chemical, bactericidal, and fungicidal properties of treated water, and explains the prolonged microbial resistance of water (PMRW)—the phenomenon when bacteria continue to be destroyed for a long time (several months) after treatment by PED. The results of investigations make it possible to conclude that the PMRW is caused by the cooperative bactericidal action on microorganisms of both oxide nanoparticles of electrodes metal and positive ions emitted by the nanoparticles.

**Index Terms**—Bactericidity, microbial resistance, nanoparticles, pulsed electric discharges (PEDs), water.

## I. INTRODUCTION

THE MEDICAL investigations for the last decades have shown that the sharp rise of oncological and cardiovascular diseases results from a chlorine disinfection of water [1]–[3]. It was revealed that chlorinated potable water contains steady macroradicals, which provoke occurrence and development of those and other serious diseases [4].

This determines an importance and urgency of work, which directed on searches of the alternative and safe ways of water disinfection. Moreover, studies of pulsed electric fields' influence on man's cells and organic tissue, including cancer tissue [5], are currently being implemented.

There are some methods for water and air purification from microbic and other pollutions which use the electric energy, the most effective are pulse electric fields [6], [7], glow discharge [8], [9], streamer/corona discharge [10], [11], gliding discharge [12], barrier discharge [13], and pulse spark discharge [14]–[17].

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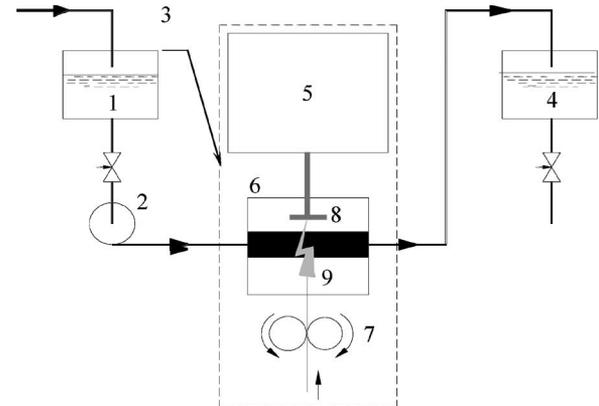


Fig. 1. Apparatus for electrodischarge treatment of water. 1—water to be treated; 2—pump; 3—unit for water treatment; 4—treated water; 5—generator of electric pulses; 6—discharge chamber; 7—feeder of wire; 8—plate; and 9—wire.

All methods aforementioned are of approximately equal efficiency for microbes' destruction because, in all those cases, the acting factors are almost the same: UV radiation, OH radicals,  $H_2O_2$ ,  $O_3$ , etc. However, pulsed electric discharges (PEDs) have essential advantages in comparison with the others: first, intensive shock waves and, second, the effect of prolonged microbial resistance of water (PMRW). Due to the latter property, water treated by PED is the bactericidal agent.

Although investigations of PED, electric fields, corona discharges, and other methods of water disinfection were carried out for more than 30 years, in this area, unresolved problems still exist, in particular, the origin of PMRW phenomenon [18], [19].

When the confirmation of absence of negative action of the electric methods on human beings was obtained, their application in medicine, pharmaceuticals, food industry, and other areas is widely expected [20], [21]. Currently, those electric methods in disinfecting the drains of hospitals and industrial enterprises can be used.

Thus, the goal of this investigation is in defining the opportunities of PED for safer disinfecting of water and other possible applications.

## II. EXPERIMENTAL SETUP AND PARAMETERS OF PED

Fig. 1 shows the diagram of apparatus for electric-discharge treatment of water.

The apparatus provides the continuous operation for months due to feeder (7), which keeps the interelectrode distance

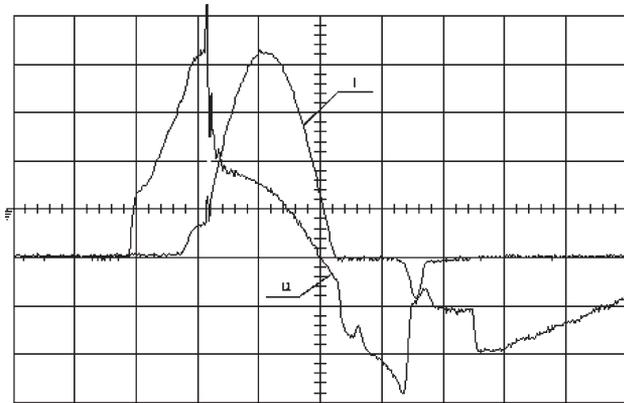


Fig. 2. Oscillograms of the current and voltage of the discharge pulse. I—current (10 A/div), U—voltage (400 V/div), time—10  $\mu$ s/div.

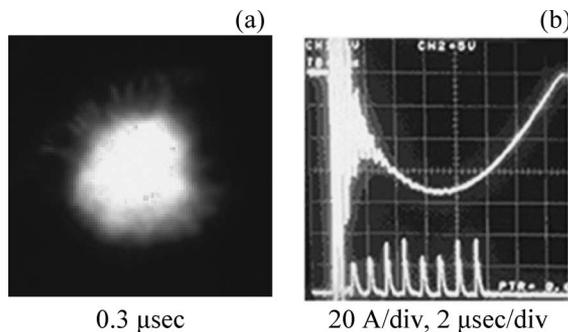


Fig. 3. (a) Glowing area at early stage of discharge and (b) discharge current.

constant by means of supplying wire (9) into the discharge chamber (6). It is necessary to compensate the increase of interelectrode gap because of electric erosion of electrodes metal.

Investigation of PED parameters, together with the physical, chemical, and bactericidal properties of treated water, has shown that the optimal PED's parameters are the following: duration: 1–20  $\mu$ s; energy: 0.2–1.0 J/pulse; current rise rate:  $10^6$ – $10^9$  A/s; and pulses frequency: 50–100 Hz [22]. The types of electrode system are “wire to plate” or “wire to wire” at the interelectrode gap of  $\sim$ 10 mm and at diameter of the channel for water passage of  $\sim$ 10 mm. The maximum bactericidal action of treated water is obtained with the electrodes made of silver, copper, or their alloys.

In Fig. 2, the typical oscillograms of the current and voltage at pulse duration of 20  $\mu$ s are presented.

Fig. 2 shows that, before the breakdown of the interelectrode gap during 5  $\mu$ s, the current increases up to 7 A and voltage up to 35 kV (the signal of voltage was flattened). After the breakdown during the discharge-column formation, the current increases up to its maximum 43 A, and the voltage, at this time, decreases from  $\sim$ 1 kV to zero at its average magnitude of  $\sim$ 700 V. The energy of a single pulse is  $\sim$ 0.4 J, and power is  $\sim$ 20 kW.

It is known that the widening of the discharge column during the early stage results in generation of a shock wave [23]–[26], behind the front of which the cavitation bubbles are formed [Fig. 3(a)] [27].

In our investigation, the velocity of the border of glowing area (from our point of view, the velocity of the shock wave) had

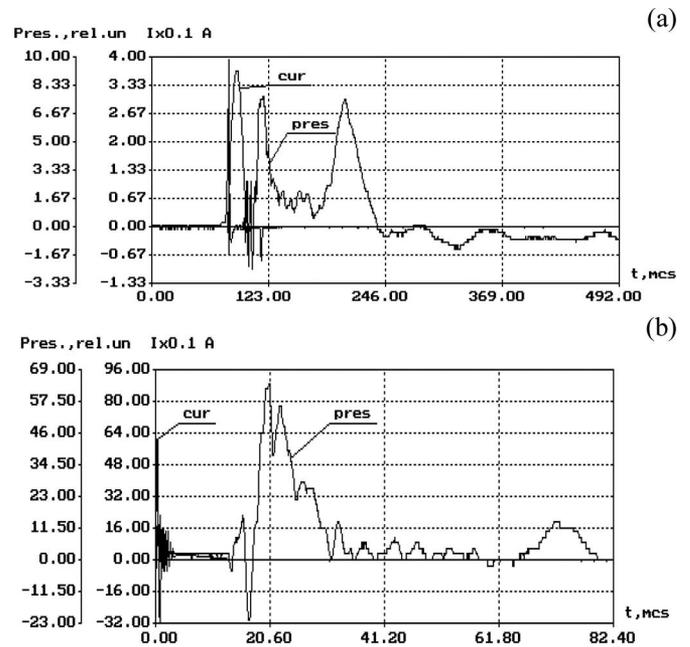


Fig. 4. Oscillograms of a current and pressure at the (a) “slow” and (b) “fast” discharges.

been registered for the time that is smaller than 0.3  $\mu$ s, current rise rate of  $5 \times 10^6$  A/s, and input energy of 0.2 J/pulse. The velocity is more than  $5 \times 10^3$  m/s [28].

Essentially lower ( $\leq$  1500 m/s) velocities of the shock-wave propagation [23], [24] are explained by high decrement of shock-wave velocity. Moreover, the measurements in those two works were carried out at the later stage of process than in our case, i.e., for the time significantly that is larger than 1  $\mu$ s [24] or at distance that is significantly larger than 10 mm from the plasma channel [23].

The shock waves in water have high gradient of pressure at their fronts, so, at the size of the bacterium, the difference of pressure is enough to damage bacterial membrane and to destroy it [29].

Comparison of pulsed pressure of the same energy of pulses for two kinds of electric discharges: “slow” at duration  $t = 20 \mu$ s,  $dI/dt = 5 \times 10^6$  A/s and “fast”  $t = 1 \mu$ s,  $dI/dt = 2 \times 10^9$  A/s. [Fig. 4(a) and (b)] shows that the increase of discharge power results in proportional increase of pulsed pressure. Therefore, for the “slow” discharges, the amplitude of shock wave measured at the wall of discharge chamber is  $\sim$ 0.5 MPa and for the “fast” ones  $\sim$ 4 MPa. Hence, the destroying effect on bacteria of the “fast” discharges is higher than of the “slow” ones at identical energy of the pulses [30].

To determine the temperature of plasma, the registration of discharge spectra by diffraction spectrometer was carried out. Fig. 5 shows the distribution of relative intensity of the discharge-column radiation at electrodes of 70% silver and 30% copper alloy.

The temperature of the discharge column, which was estimated by comparison of relative intensities of radiation in the lines, is  $\sim 10^4$  K. At that temperature, the discharge column is a source of the UV radiation in a wide wave range, in particular, 200–400 nm [31], [32]. The UV radiation, being absorbed by

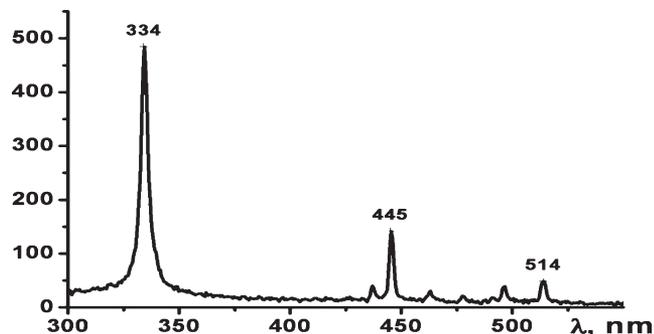


Fig. 5. Spectrum of the discharge radiation (intensity in relative units).

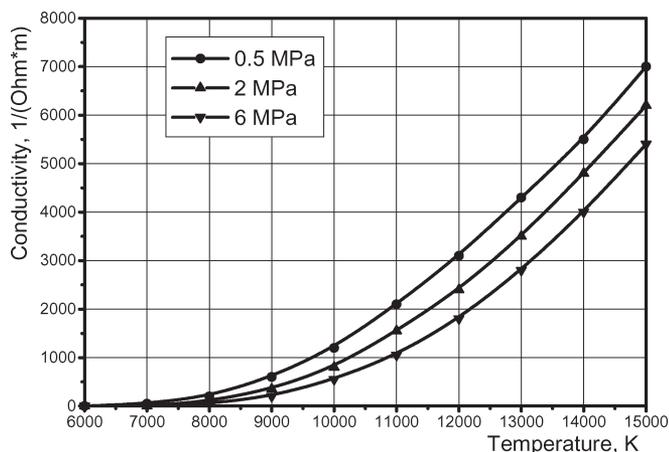


Fig. 6. Specific electroconductivity of plasma as a function of its temperature and pressure.

water, produces  $H_2O_2$ ,  $O_3$ , and OH radicals [33], [34], which destroy microbes and also some organic compounds.

The average magnitude of specific electric conductivity is  $\sim 10^3 (\Omega \times m)^{-1}$ . It was estimated for the following parameters of the discharge column: diameter  $\sim 1$  mm, length 10–13 mm, and resistance 15–20  $\Omega$ .

Fig. 6 shows the plasma electric conductivity as a function of its temperature and pressure, which were calculated under assumption of thermodynamic-equilibrium structure.

Thus, from Fig. 6, the temperature of the discharge column at pressures of 0.5–6 MPa is  $(0.9\text{--}1.1) \times 10^4$  K that is in accordance with the data above.

### III. ELECTRODE EROSION AND NANOPARTICLES

To find out the mechanism of PMRW, the physical and chemical properties and bactericidal activity of water treated by PED were investigated.

At the initial stage of investigation, the electrodes of various metals and their alloys were used. Fig. 7 shows the specific erosion of some of those metals and alloys.

We used obtained data for roughly estimating the total mass strength of the nanoparticles in the water. Furthermore, after the connection between the properties of electrode materials and bactericidal action of the treated water was found, the investigations were concentrated on three metals: silver, copper, and iron. The first two metals were chosen because their ions have the highest toxicity to bacteria and, so, are useful for

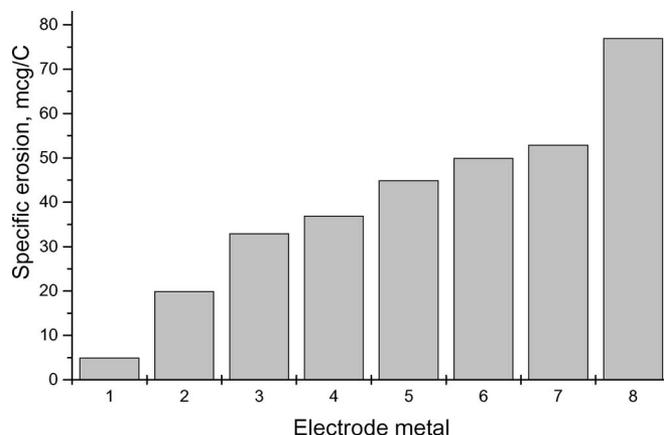


Fig. 7. Specific erosion of electrode materials. 1—W + Cu; 2— $Cr_3C_2$  + Cu; 3—Ag + Cu; 4—Fe; 5—Mo + W + Cu; 6—W + Ni + Fe; 7—Cu; and 8—W + Ni + Cu.

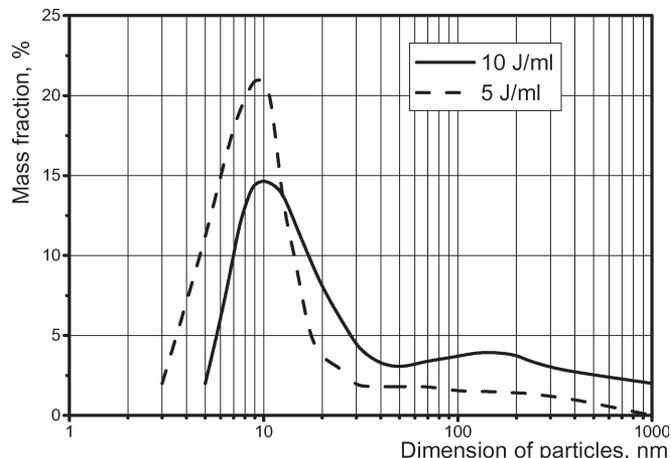


Fig. 8. Distributions in the dimension of the average values of mass fractions of silver, copper, and iron particles.

practical applications, and iron was chosen for comparison with the first two metals.

It was confirmed by the mass spectrometry with inductively connected plasma that the electrodes' erosion produces particles of metal in water. Consequently, the “Coulter N4” analyzer of submicrometer particles size was used for determination of the distributions of the particles mass fractions in size. Fig. 8 shows plots of distributions in size of the average values of mass fractions of silver, copper, and iron particles at input energy of 5 and 10 J/ml.

The plot shows that all particles are ranged from 5 up to 1000 nm, and the maximal mass fraction is of “small” particles of about 10 nm in size (hereafter nanoparticles), and that the mass fraction of these nanoparticles decreases with increase of input-specific energy.

Fig. 9 shows electronic microscopy photos of silver [Fig. 9(a)] and copper [Fig. 9(b)] nanoparticles and the clusters they have formed.

Investigation by the exclusive liquid chromatography, electronic paramagnetic resonance, spectroscopy in UV, and visible spectra of water treated by copper electrodes shows that the material of nanoparticles is not metal but its oxides of  $Cu_2O$  and  $CuO$ .

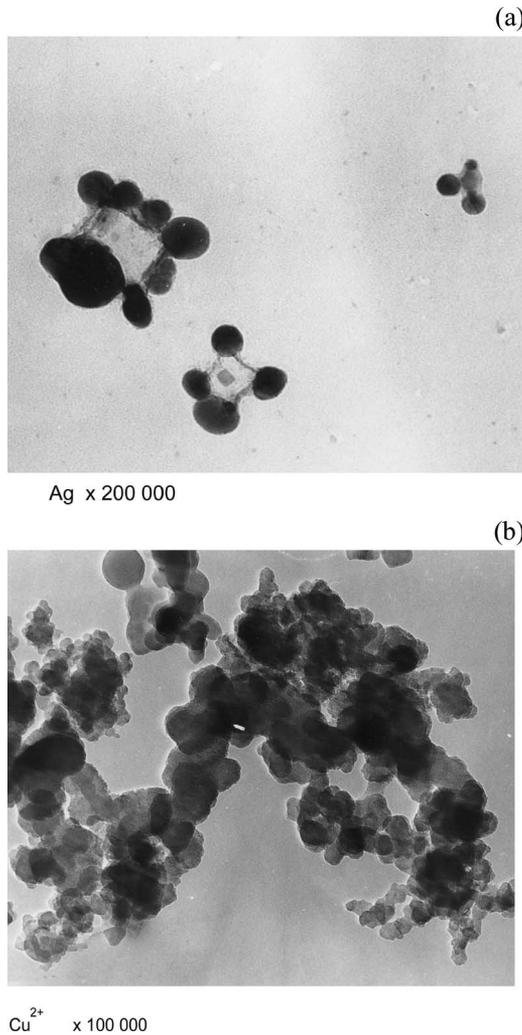


Fig. 9. (a) Silver and (b) copper nanoparticles.

Thus, it was found that the water treated by PED at copper electrodes is a dispersion containing both nanoparticles of electrodes metal and ions  $\text{Cu}^+$  and  $\text{Cu}^{++}$ , which are in dynamic concentration equilibrium [35].

After the dispersion, the nanoparticles have been extracted and then put in the deionized water; about 1 h later, it was detected that the ion concentration reached the initial level, hence, the conclusion can be made that the oxide nanoparticles are sources of ions.

The mass strength of silver, copper, and iron nanoparticles in a dispersion versus input energy at complete decomposition of the nanoparticles to the ions at  $\text{pH} = 2.5$  by means of the chromatography had been determined (Fig. 10).

By capillary electrophoreses, it was also found that the nanoparticles have a negative specific surface electric charge of  $(0.4-1.6) \times 10^{-2} \text{ C/m}^2$  and volume specific charge of  $(0.01-2.6) \times 10^{-2} \text{ C/ml}$ .

#### IV. BACTERICIDAL ACTION OF WATER TREATED BY PED

As explained above, the bactericidal activity of water treated by PED depends on the input energy and the properties of the metal electrodes. The bactericidal activity of water was deter-

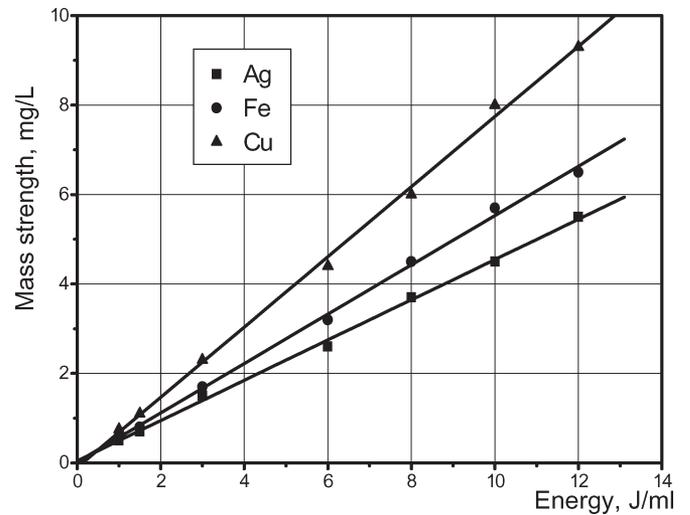


Fig. 10. Mass strength of silver, copper, and iron nanoparticles in dispersion as a function of energy input in the discharge.

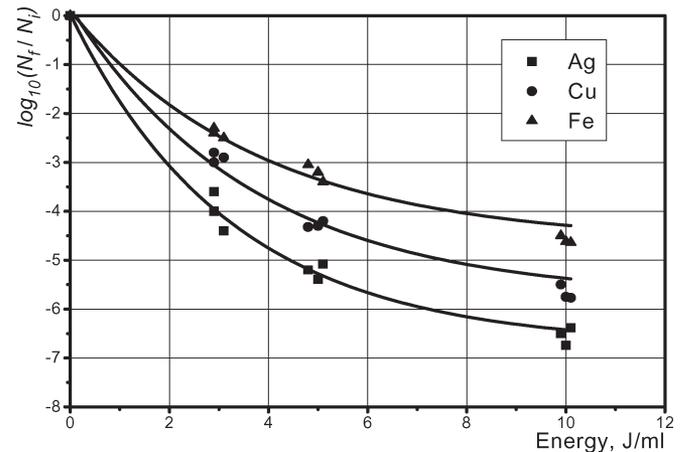


Fig. 11.  $\log_{10}(N_f/N_i)$  versus the input energy for *E. coli*.

mined as  $\log_{10}(N_f/N_i)$ , where  $N_i$  is the initial concentration of bacteria and  $N_f$  is the final concentration of viable bacteria after PED treatment, as shown in Fig. 11.

Plots show that the water treated by silver electrodes has the highest bactericidal activity in spite of mass strength of iron and copper that is higher than that of silver at identical input energy (Fig. 10).

This is because the ions of silver have the highest toxicity to bacteria. The ions of metals are arranged in the following order in terms of their toxicity:  $\text{Ag} > \text{Cu} > \text{Cd}, \text{Zn}, \text{Pb} > \text{Mn}, \text{Fe} > \text{Mg}, \text{Ca}$  [36].

Since both the mass strength of nanoparticles and bactericidal activity of the treated water are proportional to the input energy, there is also a connection between bactericidal activity and mass strength of nanoparticles. Moreover, bactericidal activity is proportional to the mass strength of nanoparticles.

The antimicrobial activity of water has also been examined with *Klebsiella pneumonia*, *Staphylococcus aureus*, *Pseudomonas aeruginosa*, *Salmonella typhimurium*, *Serratia marcescens*, *Citrobacter freundii*, *Bacillus subtilis*, *Candida albicans*, and *Ulocladium chartarum*. In all those tests, we have obtained similar results.

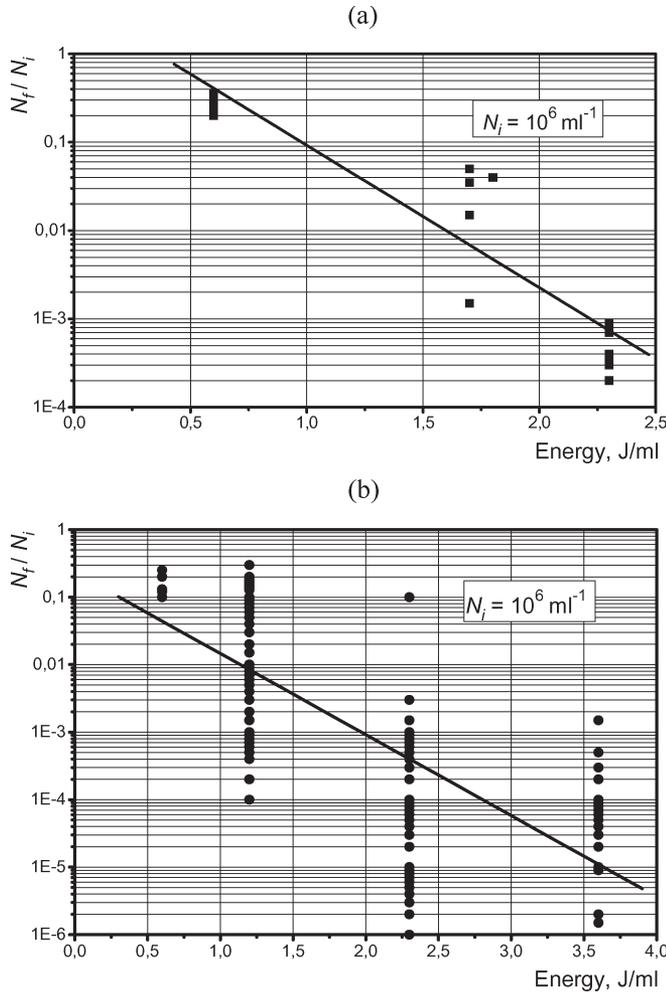


Fig. 12. Ratio of final concentration  $N_f$  of viable *E. coli* to their initial concentration  $N_i$  as a function of input energy at (a) one chamber and (b) four chambers.

Fig. 12 shows the ratio of final concentration  $N_f$  of viable *E. coli* to their initial concentration  $N_i$  ( $10^6 \text{ ml}^{-1}$ ) as a function of input energy, which is obtained with the apparatus equipped with one or four discharge chambers, connected serially by water current.

It was found that the PED treatment at the same input energy of water by means of several discharge chambers connected this way is more effective than by one chamber (Fig. 12). Comparison of plots (Fig. 12) shows that the final concentration  $N_f$  of viable *E. coli* to their initial concentration  $N_i$  ( $10^6 \text{ ml}^{-1}$ ) at input energy of 2.5 J/ml is decreased from  $3 \times 10^{-3} \text{ ml}^{-1}$  at one discharge chamber up to  $10^{-4} \text{ ml}^{-1}$  at four chambers.

This difference may be explained by multiple actions of shock waves on bacteria and mixing of treated water in the second case.

Since the treated water can be considered as the bactericidal agent, it was necessary to determine its bactericidal activity as a function of a ratio of dilution of the treated water by the initial one.

Fig. 13 shows the final concentration of *E. coli* versus the ratio of dilution, at the input energy of 10 J/ml, at the initial concentration of bacteria of  $7.9 \times 10^3 \text{ ml}^{-1}$ , and at time of exposition of 24 h.

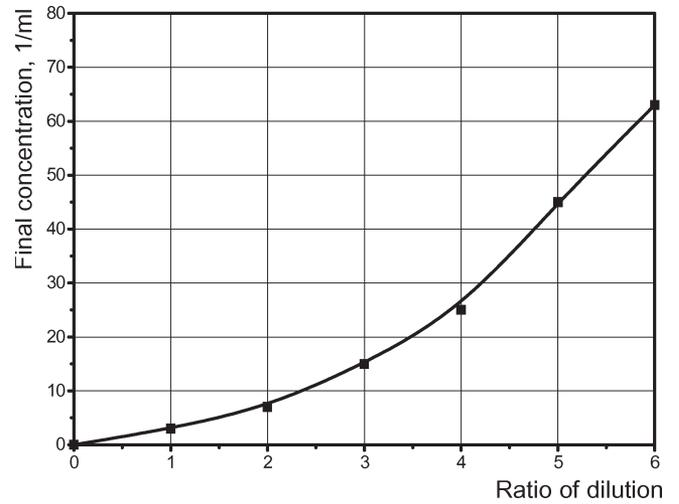


Fig. 13. Final concentration of *E. coli* versus ratio of dilution.

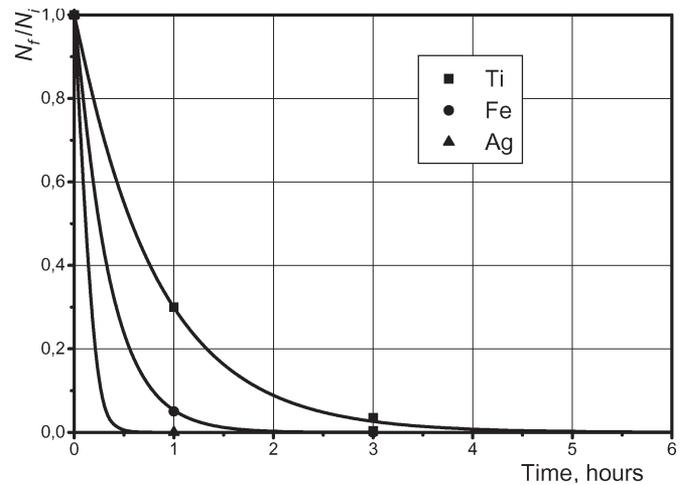


Fig. 14. Rate of destruction of the spores *U. chartarum* at the input energy of 10 J/ml.

The rate of destruction of fungi *U. chartarum* spores treated by PED titanium, iron, and silver electrodes and then exposed in treated water is shown in Fig. 14 [37].

The functions are exponential, and in a period from 15 min to 3 h, all spores were destroyed.

The rate of destruction of *E. coli* in the water treated by copper electrodes, at the input energy of 10 J/ml and the initial concentration of bacteria of  $3,8 \times 10^3 \text{ ml}^{-1}$  and  $5,1 \times 10^6 \text{ ml}^{-1}$  is presented in Fig. 15.

The functions are also exponential, and in 5 min, in dependence on the initial concentration of bacteria, from 80% up to 100% of them had been destroyed.

## V. MECHANISMS OF PMRW

It has been shown above that two groups of factors cause bactericidal action of PED in water: 1) UV radiation and shock waves, which act at discharge time—factors of current action, and 2) hydrated electrons, OH radicals,  $\text{H}_2\text{O}_2$ , nanoparticles, and positive ions of metal electrodes, which act after the treatment—factors of post action.

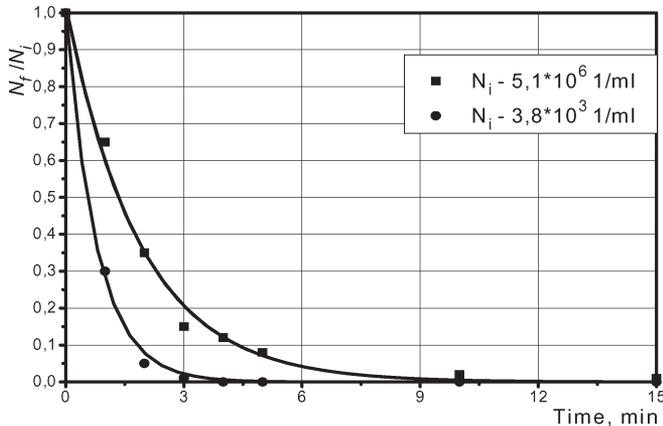


Fig. 15. Rate of destruction of *E.coli* at the input energy of 10 J/ml.

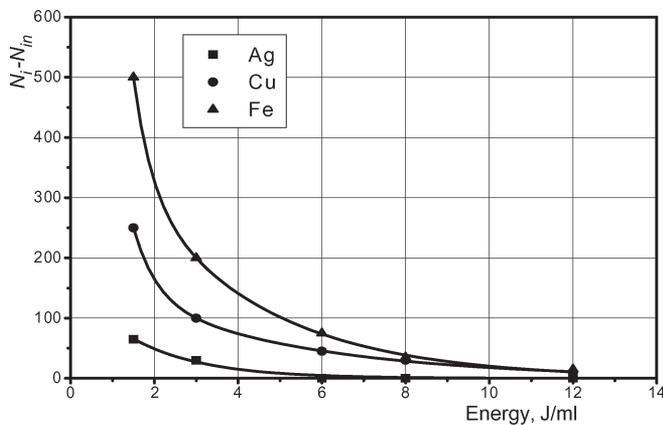


Fig. 16. Differences of final concentrations of viable spores in two kinds of treated water as a function of input energy, where  $N_i$ —containing only ions and  $N_{in}$ —containing both ions and nanoparticles.

Since hydrated electrons exist  $\sim 0.5$  ms and OH radicals and  $H_2O_2$  of no more than several days, it is evidently that they cannot be considered as factors causing the PMRW, which is of several months duration.

Thus, only the nanoparticles and positive ions of metal, which are produced by electrodes erosion, are responsible for PMRW.

To reveal the role of the nanoparticles in PMRW, experiments were carried out on inhibition of growth of *U.chartarum* fungi spores in two modifications of the same treated water: 1) containing both ions and nanoparticles and 2) containing only ions (after the removal of nanoparticles). The water was treated by means of silver, copper, and iron electrodes at specific input energy of 1.5, 3, 6, 8, and 12 J/ml.

Fig. 16 shows the differences of the final concentrations of *U.chartarum* fungi viable spores as a function of input energy, where  $N_i$  is the final concentration of spores in water containing, after the removal of nanoparticles, only ions and  $N_{in}$  is the final concentration of spores in water containing both ions and nanoparticles.

Plots of differences between the final concentrations of  $N_i$  and  $N_{in}$  at initial concentration of spores of about  $10^3$  ml $^{-1}$  after ten days incubation show that fungicide effect is higher when both ions and nanoparticles are in water. The differences

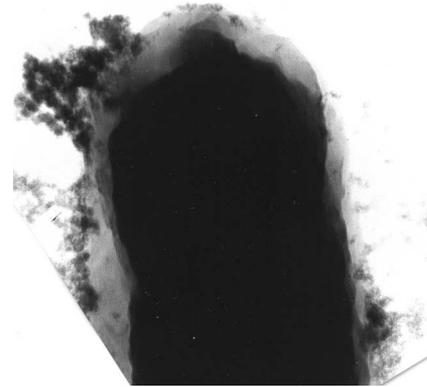


Fig. 17. Nanoparticles of Cu on the cell wall of *E.coli* ( $\times 10^5$ ).

are maximal when the material of electrodes is iron, i.e., the metal with the lowest toxicity of ions. In this case, the bactericidal effect of nanoparticles themselves is highest, whereas, at higher toxicity of ions, this difference is lower. Besides, these differences decrease at the increase of input energy, i.e., at the increase of ions and nanoparticles concentration. Thus, the nanoparticles in water increase its bactericidal action. This may be explained by two reasons: Nanoparticles, at the time of incubation, maintain the concentration of ions at the same level and the nanoparticles themselves (by means of their surface electric charge) participate in killing the spores.

The most probable mechanisms of bactericidal action of the nanoparticles on bacteria are penetration of nanoparticles inside bacteria [38], cooperative action of toxic ions emitted by nanoparticles, and surface electric charge of nanoparticles [39].

By electronic microscopy, it was detected that the single nanoparticles and their clusters settle down on bacteria cell walls.

Fig. 17 shows, in contrast to the data presented in paper [38], that nanoparticles do not penetrate inside the bacteria, but, adsorbing the ions, remove the concentration balance between ions and nanoparticles that causes additional emission of ions from nanoparticles. Because of that, when nanoparticles approach close to bacteria, directed streams of toxic ions appear, which produce the bactericidal effect.

## VI. CONCLUSION

The PED in water destroy the bacteria including the shock waves. It was measured that the velocity of their expansion, in the very early stage of discharge, is  $\geq 5 \times 10^3$  m/s.

It was found that water treated by PED is the dispersion containing both nanoparticles of metal electrodes and positive ions, which are in dynamic concentration equilibrium for several months. The particles of  $\sim 10$  nm in size have maximal subfraction.

It was shown that, at equal concentration of ions in two kinds of dispersions, the bactericidal action of dispersion, which contains both ions and nanoparticles, is higher than the one containing only ions.

Electronic microscopy showed that nanoparticles and clusters do not penetrate inside the bacteria but settle down on their

cell walls and produce streams of the toxic ions, which destroy the bacteria.

On the base of all obtained data, we concluded that the PMRW is the cooperative bactericidal action on bacteria of both oxide nanoparticles and positive ions emitting by them.

The importance of this investigation is that the obtained results connect the PMRW with the parameters of electric discharges and with the properties of metal electrodes; moreover, the results give the opportunity to produce the dispersions with the required bactericidal properties, which can be considered as the bactericidal agents.

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