

NUMERICAL INVESTIGATION OF THE INFLUENCE PRODUCED BY ELECTRIC CIRCUIT PARAMETERS ON THE FORMATION OF CHEMICALLY ACTIVE RADICALS IN WATER VAPOURS

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The numerical investigation of the formation of chemically active radicals during the discharge in water vapors has been carried out. An increase in the number of radicals in terms of time and a change in thermodynamical discharge parameters were determined for different U , L , C values of the discharge circuit. Consideration was given to the two domains, in particular the discharge channel and the region between the discharge channel and the shock wave front. Special attention was given to such chemical components as: OH , O , H , H_2O_2 , HO_2 , H_2O .

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1. INTRODUCTION

At the present time the underwater spark discharge finds many practical applications. The electrical hydraulics repeatedly makes itself known as a new chapter of electrochemistry, which is called the pulsed electrochemistry. The electrohydraulic method can be used as an alternative of chemical methods used for the treatment of the medium in such processes as the removal of incrusting matters and diamond cakes or metal catalysts from the flax fiber, and also bleaching of flax cotton wool, drinking water disinfection and waste water treatment. Therefore, a lot of experimental and theoretical work is done at the moment [1-10] to investigate the mechanisms of changes in physical and chemical properties of water and water solutions of organic and inorganic substances subjected to the action of electrical discharge. The electrohydraulic discharge has many advantages as for its chemical action on liquid media in comparison with the atmospheric pressure barrier glow discharge with the electrolytic cathode, and the spark electric air discharge in the presence of water vapors [11]. In the case of electrohydraulic discharge active radicals are formed directly in the treated medium, and this provides the volumetric treatment of the medium. Other types of discharges provide only the surface treatment that reduces their efficiency. In contrast to gas and transient discharges electric water discharges practically exclude the formation of the broad spectrum of nitrides of high toxicity and sufficiently long lifetime. A hydroxyl radical OH is rather easily formed in water discharges and it has better oxidation properties

in comparison with ozone. It should be noted that it is difficult to produce ozone in high humidity conditions because of the increased velocity of its decomposition. In previous papers consideration was given to the underwater spark discharge and its potential application for different technological processes [12-14]. These scientific papers describe the vapor-gas-plasma cavity (VGPC) in which plasma, chemically active elements and their components capable of producing influence on the acidity of treated medium are formed.

The VGPC envelope coincides with the shock wave front and it functions as an interface between the untreated liquid and the vapor-gas-plasma mixture. We gave no consideration to the availability of radicals outside the VGPC envelope. The processes that occur within the VGPC envelope require more detailed studies to reveal the areas of accrual of some chemically active radicals, for example, OH , HO_2 those that are capable of changing the acidity of the treated medium. There are many mathematical models [15-18] that allow for the adequate description of the processes that occur in the electrohydraulic discharge. However, the chemical composition of the medium for the known compositions is calculated on the assumption of the establishment of chemical equilibrium. The characteristic times of the behavior of chemical processes at abrupt discharges can exceed the characteristic times of the behavior of electrophysical processes. Therefore we need to use mathematical models that take into consideration nonequilibrium chemical kinetics and allow for

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the estimation of formed active radicals. A scientific paper [19] proposes the numerical model that takes into consideration the nonequilibrium chemical kinetics with the formation of active particles in the water exposed to the action of pulsed ultraviolet radiation created over the liquid surface by the independent spark discharge. The model [19] allows for the determination of a change in the concentration of primary products that are formed in water. However, the mechanism of a change in physical and mechanical properties of water and water solutions at the electrohydraulic discharge differs from that of spark discharges over the liquid surface. This prevents us from using the known model [19] to study the processes in the electrohydraulic discharge. Due to the fact that it is rather difficult to take into consideration the liquid resistance to the dynamics of the VGPC boundary for the numerical simulation, the first stage of the research deals with the studies of the influence the parameters of electrical circuit on the formation of chemically active radicals in water vapors.

2. DESCRIPTION OF THE MODEL AND SIMULATION CONDITIONS

For the numerical simulation of the spark discharge in water vapors we used the mathematical model that was described in detail in scientific paper [19] and that takes into consideration the nonequilibrium chemical kinetics with changed initial simulation conditions. The spark channel expansion process was described by the following equation system:

$$\frac{\partial \vec{a}}{\partial t} + \frac{1}{r} \frac{\partial r \vec{b}}{\partial r} = \frac{1}{r} \vec{f}, \quad (1)$$

Where the column vectors are equal to:

$$\begin{aligned} \vec{a} &= \begin{pmatrix} \rho \\ \rho u \\ \rho \varepsilon + \rho u^2 / 2 \\ y_i \end{pmatrix}, \\ \vec{b} &= \begin{pmatrix} \rho u \\ p + \rho u^2 \\ u(\rho \varepsilon + \rho u^2 / 2 + p) + k_T dT/dr \\ u y_i \end{pmatrix}, \\ \vec{f} &= \begin{pmatrix} 0 \\ p \\ r(\sigma E^2 - Q_{em}) \\ r \dot{\omega}_i \end{pmatrix}, \end{aligned} \quad (2)$$

where ρ is the gas density, u is the velocity, p is the pressure, ε is the gas internal energy per gas mass unit, k_T is the heat conduction coefficient, E is the electric field intensity in the discharge channel column, σ is the channel plasma conductivity, Q_{em} is the discharge energy losses for the radiation including the ionization and dissociation, r is the radius coordinate, T is the gas temperature, y_i is the molar concentration of the i -th component (OH , O , H , H_2O_2 , HO_2 , H_2O); is the rate of change in the concentration

of the i -th component of the mixture due to chemical reactions. The model assumes that the local thermodynamic equilibrium is established in the domain of the current-carrying channel. The processes that occur outside the current-carrying channel were calculated using the equations of nonequilibrium chemical kinetics [20]. Electric processes in the discharge circuit were expressed by the equation

$$L \frac{di}{dt} + [R_c + R_s(t)]i + \frac{1}{C} \int_0^t i dt = U_0, \quad (3)$$

where L is the equivalent inductance of the discharge circuit, i is a current of the discharge circuit, R_c is the active resistance of the discharge circuit, R_s is the resistance of the spark, C is the electric capacitance of the capacitor, U_0 is the initial voltage of the capacitor charge. The formation of chemically active radicals was studied in compliance with chemical reactions (Tab.1).

Table 1. A simplified kinetic circuit of the behavior of chemical reactions [21] adopted for the model (M denotes the third particle)

| Number | Reactions |
|--------|--|
| 1 | $H_2O + M \leftrightarrow H + OH + M$ |
| 2 | $H_2O + H \leftrightarrow OH + H_2$ |
| 3 | $H + O_2 + O_2 \leftrightarrow HO_2 + O_2$ |
| 4 | $H + O_2 + H_2O \leftrightarrow HO_2 + H_2O$ |
| 5 | $H + O_2 \leftrightarrow O + OH$ |
| 6 | $H + HO_2 \leftrightarrow O_2 + H_2$ |
| 7 | $H + HO_2 \leftrightarrow OH + OH$ |
| 8 | $H + H_2O_2 \leftrightarrow HO_2 + H_2$ |
| 9 | $H + O_2 + M \leftrightarrow HO_2 + M$ |
| 10 | $OH + OH + M \leftrightarrow H_2O_2 + M$ |
| 11 | $OH + HO_2 \leftrightarrow O_2 + H_2O$ |
| 12 | $OH + H_2O_2 \leftrightarrow HO_2 + H_2O$ |
| 13 | $HO_2 + HO_2 \leftrightarrow O_2 + H_2O_2$ |
| 14 | $O + O + M \leftrightarrow O_2 + M$ |
| 15 | $O + H + M \leftrightarrow OH + M$ |
| 16 | $H + OH \leftrightarrow O + H_2$ |
| 17 | $H + H + M \leftrightarrow H_2 + M$ |

For the initial conditions in the calculated region we specified the availability of water vapors with the following thermodynamic parameters:

$$p(r)|_{t=0} = p_0, \quad T(r)|_{t=0} = T_0, \quad u(r)|_{t=0} = 0, \quad (4)$$

where $p_0 = 10^5 Pa$, $T_0 = 473 K$. The given temperature and pressure provide the condition for the occupation of the calculated area with water vapors. The resistance of the discharge circuit was assumed to be equal to $R_c = 0.1 Ohm$. The influence produced by the parameters of electric circuit on the formation of radicals was studied by varying the value of one of the parameters (C , U_0 , L). The values of the parameters are $C = \{2\mu F, 4\mu F, 6\mu F\}$,

$U_0 = \{15 \text{ kV}, 20 \text{ kV}, 25 \text{ kV}\}$, $L\{3 \mu\text{H}, 5 \mu\text{H}, 7 \mu\text{H}\}$. The length of the discharge gap was assumed to be equal to $l = 5 \text{ mm}$. For the boundary conditions it was assumed that the gradients of thermodynamic parameters on the discharge channel axis are equal to zero. The processes of the intensive nucleation of chemically active radicals occur in the first quarter of the period of oscillatory decaying discharge [21]. Therefore, the calculated data are given for this particular period of time. It is known [22] that the chemical activation region is limited during the discharge by the parameter $B = 4l$, where l is the length of the interelectrode gap. Therefore the size of the calculated region was The equation system of the gas dynamics (1) was calculated using the S.K. Godunov scheme [23].

3. MATHEMATICAL SIMULATION DATA

To study the general mechanisms of the change in the chemical composition of the discharge medium it is necessary to consider the kinetics of the change of the mole concentrations of the components along the radius of the spark channel with the elapse of time (Figs.1,a-c and Fig.2). Water dissociation into hydrogen and oxygen occurs in the spark channel under the action of high temperature. The concentration of water vapors increases while moving away from the discharge channel axis due to the drop in the medium temperature (see Fig.2). The concentration peak of water vapors is in the middle of the region between the discharge channel and the shock wave front, which is related to the medium compression process after the shock wave front at the gas dynamic expansion of the spark channel.

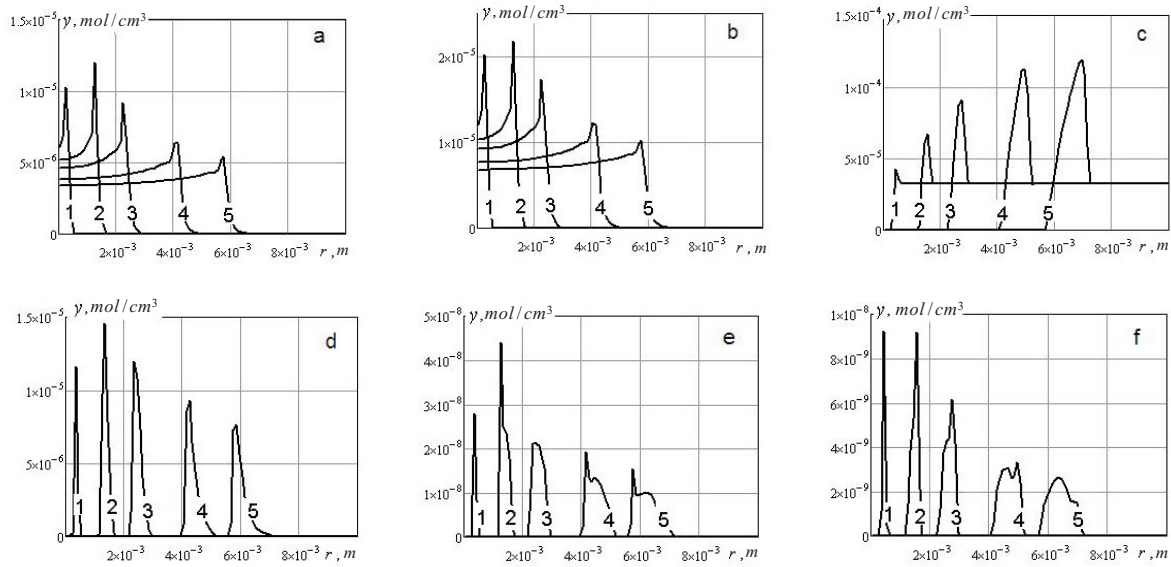


Fig.1. The distribution of the mole concentrations of components as a function of the distance along the radius [a) — O ; b) — H ; c) — H_2O ; d) — OH ; e) — HO_2 ; f) — H_2O_2] across the channel section at a time point of 1...0.1 μs , 2...0.5 μs , 3...1 μs , 4...2 μs , 5...3 μs (the discharge at $C = 2 \mu\text{F}$, $U = 25 \text{ kV}$, $L = 3 \mu\text{H}$)

Figs.1,a-c show that all chemical components, except water are characterized by a decrease in their maximum concentration values after 0.5 μs from the beginning of the discharge progress. The radicals O and H are formed not only in the current carrying channel but also in the region adjacent to the channel and limited by the shock wave front. An increase in the number of radicals is caused by nonequilibrium chemical reactions. The radicals OH , HO_2 , and peroxide H_2O_2 are formed in the region between the discharge channel and the shock wave front (Figs.1,g,d,e). In the region of the current-carrying channel the concentration of the components OH , HO_2 , and peroxide H_2O_2 is negligibly small (see Fig.2). Therefore, an increase in the concentration of the given radicals begins from the moment of the separation of the shock wave front from the current-

carrying channel, which in terms of calculations corresponds to the time range of 0.2...0.5 μs . The concentration maximum occurs at a distance of 2 mm from the discharge channel axis. As the spark discharge develops the region of OH , HO_2 , and peroxide H_2O_2 formation moves away from the channel axis. As a result the radical formation space is increased in proportion to the radius square (of the current position of the formation region).

The comparative analysis of the distribution of the mole concentrations of considered active radicals along the radius at a time point of $t = 3 \mu\text{s}$ is given in Fig.2. It has been established that the O , H and OH components have the highest concentration and the concentration of HO_2 and H_2O_2 is several orders lower. This gives the grounds to suppose that a change in the chemical composition

of the medium at the water discharge can mainly be provided due to the action of the O , H and OH .

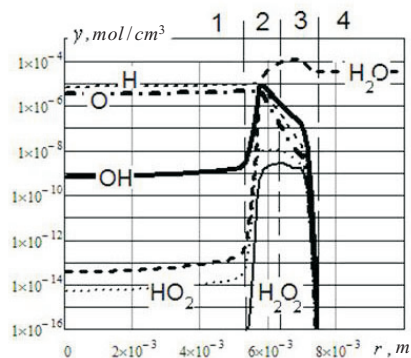


Fig.2. The distribution of the mole concentrations of the components along the radius at a time point of $t = 3 \mu s$ (for the discharge at $C = 2 \mu F$, $U = 25 kV$, $L = 3 \mu H$)

Since the conditions for the progress of chemical reactions are created between the current-carrying

channel and the shock wave front and the concentration of the OH radical in this region exceeds the concentration of other components one can state that the main physical and chemical transformations that occur at the liquid discharge are related to the formation of the hydroxyl radical OH . The vertically dotted line shows the regions under consideration. 1 — the discharge channel, 2 — the first half of the region between the discharge channel and the shock wave front, 3 — the second half of the region between the discharge channel and the shock wave front, 4 — the region in front of the shock wave front. The calculated concentrations of active radicals at a time point of $t = 3 \mu s$ are given in Table 3. It should be noted that the jump in HO_2 and H_2O_2 concentrations is the most intensive and it changes by 8 to 11 orders accordingly, while the OH concentration increases by 4 orders. The concentration of hydrogen and oxygen is somewhat lower in the current-carrying channel due to a decrease in density. However, in the region between the channel and the shock wave front the radicals O , H and OH have the highest concentration.

Table 2. The concentration of active components at a time point of $3 \mu s$ after the initiation of the spark channel in water vapors

| Active radicals | Concentration mol/cm^3 in the discharge channel Region N1 | Concentration mol/cm^3 , the first half the region between the discharge channel and the shock wave front Region N2 | Concentration mol/cm^3 , the second half of the region between the discharge channel and the shock wave front Region N3 |
|-----------------|---|---|---|
| H | $8 \times 10^{-6} \dots 10^{-5}$ | $10^{-5} \dots 7 \times 10^{-7}$ | $7 \times 10^{-7} \dots 0$ |
| O | $5 \times 10^{-6} \dots 7 \times 10^{-6}$ | $7 \times 10^{-6} \dots 10^{-7}$ | $10^{-7} \dots 0$ |
| OH | $9 \times 10^{-10} \dots 4 \times 10^{-9}$ | $4 \times 10^{-9} \dots 2 \times 10^{-6}$ | $2 \times 10^{-6} \dots 0$ |
| HO_2 | $7 \times 10^{-15} \dots 6 \times 10^{-14}$ | $6 \times 10^{-14} \dots 10^{-8}$ | $10^{-8} \dots 0$ |
| H_2O_2 | $5 \times 10^{-19} \dots 10^{-16}$ | $10^{-16} \dots 5 \times 10^{-9}$ | $5 \times 10^{-9} \dots 0$ |

Figs.3,a,b,c,d gives the results of the studies of the influence produced by electric circuit parameters on the formation of the hydroxyl radical OH , and also on the distribution of thermodynamic parameters in the spark channel. The subsequent comparative analysis of the influence of electric circuit parameters was done with regard to the calculation variant: $U_0 = 25 kV$, $C = 2 \mu F$, $L = 3 \mu H$ (see Fig.3,a). Calculated data that are given in Fig.3,b were obtained by changing the voltage U_0 . To obtain further results we changed the capacitor capacitance (see Fig.3,c) and the discharge circuit induction (see Fig.3,d). Only one circuit parameter was changed: the others remained unchanged. It has been established that as the capacitor charge voltage drops from $25 kV$ to $15 kV$, the mean temperature and the pressure in the region of the progress of nonequilibrium chemical reactions (between the current-carrying channel and the shock wave front) reduce by the time point of $3 \mu s$ from $3500 K$ to $2500 K$, and from $1.5 MPa$ to $1 MPa$,

accordingly. (see Fig. 3 a, b). The concentration of the radical OH drops from $8 mol/cm^3$ at $25 kV$ to $5, 8 mol/cm^3$ at $15 kV$. A change in the capacitance from $2 \mu F$ to $6 \mu F$ affects the OH formation to a lesser extent, though the mean values of temperature and pressure are somewhat increased (see Fig 3 a, b). The increase in the discharge circuit induction from $3 \mu H$ to $7 \mu H$ results in a decrease of the average concentration of the hydroxyl radical down to $5 mol/cm^3$ (see Fig.3,a,d). It should be noted that the change of either electric parameter results in no characteristic region for the formation of active radicals. The maximum values of the concentration of hydroxyl radicals are observed in the section that is closer to the current-carrying channel. It has thus been established that the formation of chemically active radicals, in particular the hydroxyl radical OH occurs in the region between the discharge channel and the shock wave front and directly depends on the current and thus the electric parameters of the discharge loop.

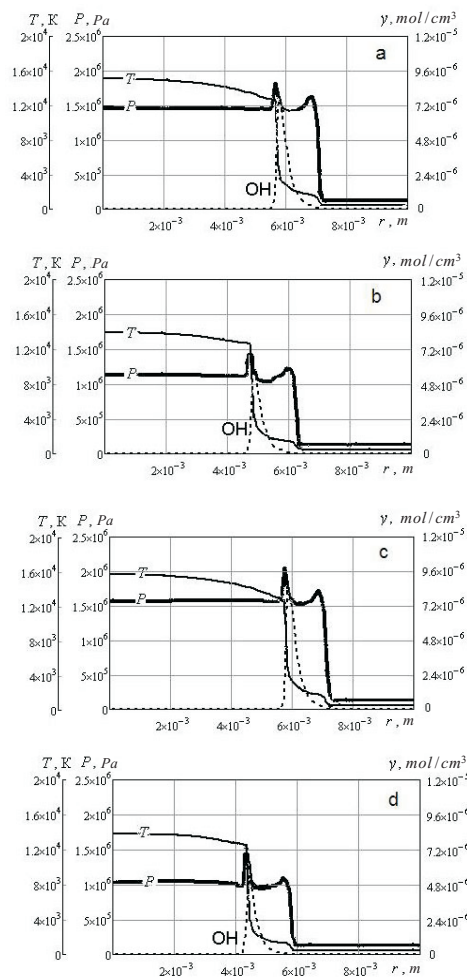


Fig.3. The distribution of the mole concentrations of the hydroxyl radical OH taking into consideration the changes in temperature and pressure as a function of a distance along the radius at a time point $t = 3 \mu s$. For a)– $U = 25 \text{ kV}$, $C = 2 \mu F$, $L = 3 \mu H$; b)– $U = 15 \text{ kV}$, $C = 2 \mu F$, $L = 3 \mu H$; c)– $U = 25 \text{ kV}$, $C = 6 \mu F$, $L = 3 \mu H$; d)– $U = 25 \text{ kV}$, $C = 2 \mu F$, $L = 7 \mu H$

Figs.4, 5, 6 show the distribution relationships of the mole concentrations of the atomic oxygen and the OH radical as a function of discharge circuit parameters. We can see that an increase in the voltage and battery capacitance results in a change of the concentrations of both radicals. An increase in the induction results on the contrary in a decrease in the formation of the radicals proposed for the consideration.

The experimental papers show that the electric field intensity affects the formation of active radicals and the intensity can be controlled in its turn using electric circuit parameters. The formation of active radicals intensifies as the intensity increases and the capacitance factor and the induction factor are optimized. The above Figures show that the realization of these relationships results in higher concentrations of the OH radical at initial discharge stages and general increase in the formed substance across the distance along the discharge channel section.

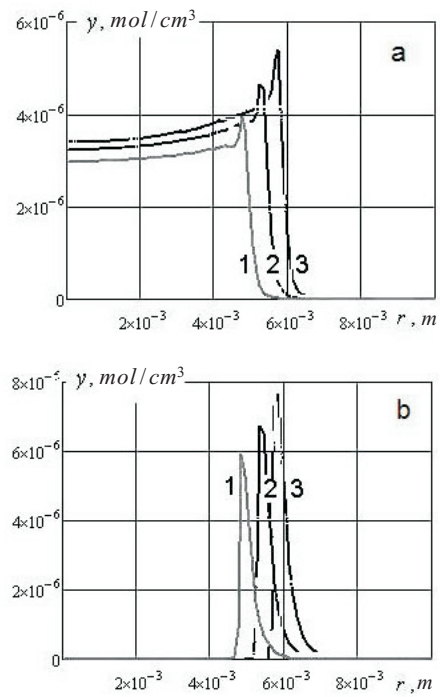


Fig.4. The distribution of the mole concentrations of the components as a function of a distance along the radius at a time point of $t = 3 \mu s$. a) – O , and b) – OH . For $C = 2 \mu F$, $L = 3 \mu H$, 1...15 kV, 2...20 kV, 3...25 kV

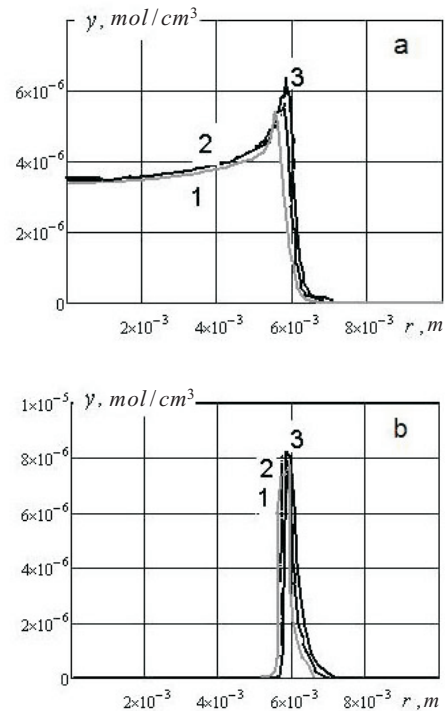


Fig.5. The distribution of the mole concentrations of the components as a function of a distance along the radius at a time point of $t = 3 \mu s$: a) – O , and b) – OH . For $U = 25 \text{ kV}$, $L = 3 \mu H$, 1...2 μF , 2...4 μF , 3...6 μF

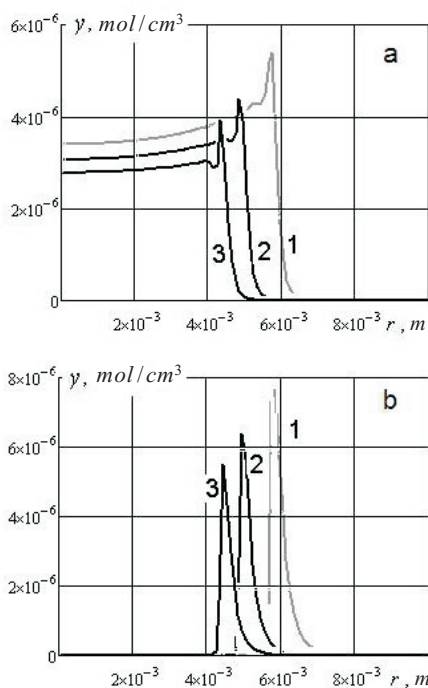


Fig. 6. The distribution of the mole concentrations of the components as a function of a distance along the radius at a time point of $t = 3 \mu s$. a) — O , and b) — OH . For $C = 2 \mu F$, $U = 25 kV$, 1... $3 \mu H$, 2... $5 \mu H$, 3... $7 \mu H$

4. CONCLUSIONS

The numerical investigation of the influence produced by the electric parameters of the discharge circuit on the formation of chemically active radicals OH , O , H_2O_2 , HO_2 at the development of the spark discharge in water vapors has been carried out. It has been established that by changing the parameter values of the discharge circuit one can regulate the amount of the produced substance and control thermodynamic parameters. The induction and voltage affect significantly the values under study. A decrease in the discharge circuit induction and an increase in the voltage result in the intensified formation of active radicals. A change in the capacitance in the considered range produces a lower influence. During the liquid discharge active particles are formed both in plasma, i.e. in the current channel and in the gas-vapor phase, i.e. the region between the outer channel boundary and the shock wave front, which corresponds to the internal space of vapor-gas-plasma cavity. Hydrogen and oxygen are prevalent in the area adjacent to discharge channel. The most active formation phase is observed in the region adjacent to the current channel. Near the internal boundary of the shock wave front the intensity of the reactions is decreased and as a consequence the formation of active radicals decays. In the region between the current channel and the shock wave front we observe an increase in the concentration of OH , H_2O_2 , HO_2 radicals with a simultaneous precocious decrease in O and H concentrations. Therefore, during the assem-

bly of electrical equipment used for electrochemical purposes, in particular for the generation of chemically active particles, or to change pH of the medium or to intensify the bactericidal effect action on the pathogenic microflora we recommend to decrease induction factors with a simultaneous increase in U and C parameters. From the point of view of the practical application the variant of the change of the charge voltage to improve radical formation factors will be the most optimal. It has been established that the hydroxyl radical OH and the atomic oxygen that define the medium acidity prevail in the given time ranges for studied radicals, which is important for different practical applications. One can assume that for practical purposes the formation of chemically active radicals in treated liquid media can be controlled using the electrohydraulic method, which in its turn can provide many advantages in comparison with some chemical methods of treatment, for example, using the chlorination.

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ЧИСЛЕННОЕ ИССЛЕДОВАНИЕ ВЛИЯНИЯ ПАРАМЕТРОВ ЭЛЕКТРИЧЕСКОЙ ЦЕПИ НА НАРАБОТКУ ХИМИЧЕСКИ АКТИВНЫХ РАДИКАЛОВ В ПАРАХ ВОДЫ

Д. В. Винников

Проведено численное исследование образования химически активных радикалов при разряде в парах воды. Для различных значений U , L , C разрядной цепи определены наработка радикалов во времени и изменение термодинамических параметров разряда. Рассмотрены две области образования радикалов: канал разряда, область между каналом разряда и фронтом ударной волны. Рассматривались химические компоненты: OH , O , H , H_2O_2 , HO_2 , H_2O .

ЧИСЕЛЬНЕ ДОСЛІДЖЕННЯ ВПЛИВУ ПАРАМЕТРІВ ЕЛЕКТРИЧНОГО КОЛА НА НАПРАЦЮВАННЯ ХІМІЧНО АКТИВНИХ РАДИКАЛІВ У ПАРАХ ВОДИ

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Проведено чисельне дослідження утворення хімічно активних радикалів при розряді в парах води. Для різних значень U , L , C розрядного кола визначено напрацювання радикалів у часі та зміна термодинамічних параметрів розряду. Розглянуті дві області утворення радикалів: канал розряду, область між каналом розряду і фронтом ударної хвилі. Розглядалися хімічні компоненти: OH , O , H , H_2O_2 , HO_2 , H_2O .