# INFLUENCE OF ULTRASONIC RADIATION ON THE PROPERTIES OF PLASMA IN A GAS DISCHARGE IMMERSED IN A LIQUID[1]

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Experimental researches of the properties of plasma–liquid systems have been carried out by studying an electrical discharge in the gas channel with a liquid wall, with additional excitation of the ultrasonic field in a liquid. The ultrasound field in the liquid phase of such systems was demonstrated to enhance the nitrous acid yield and affect the composition evolution in the plasma radiation spectrum.

## 1. Introduction

Plasma-liquid systems with an atmospheric-pressure transverse discharge can be used in such technological processes as nanoparticle synthesis, gasification of organic waste materials, and others [1–3]. A quite interesting application of similar systems to the solution of ecological problems is the reforming of liquid hydrocarbons and the destruction of stable toxic hydrocarbonic compounds in aqueous solutions. The plasma-stimulated reforming of biofuels is promising for the production of a synthetic gas enriched with hydrogen, the burning of which is accompanied by the formation of considerably less amount of harmful compounds $NO_x$–$SO_x$. The plasma-stimulated destruction of complex molecular compounds in water is rather effective for the purification of water from harmful organic pollutants.

Gas-dynamic liquid systems involving discharges in the gas channel with a liquid wall (DGCLW) [7] possess certain advantages over other plasma-liquid systems. In particular, the efficiency of the interaction between chemically active plasma components and the liquid phase of such systems is higher, because the area of the surface contact between them is larger. Moreover, the gas flow that forms the gas channel forces plasma particles to enter into the liquid, which results in an increase of the interaction time between chemically active agents formed in plasma, on the one hand, and the molecules of both the liquid itself and the substances dissolved in it, on the other hand.

It is known [8, 9] that the excitation of an ultrasonic field, the intensity of which exceeds the cavitation threshold, in a liquid exerts a promoting effect on chemical processes in solutions. The combination of such two factors as plasma and the ultrasonic stimulation of chemical processes can result in a mutual enhancement of both effects. For instance, the ultrasonic cavitation is a known mechanism of formation of micropores in a liquid. When applying a potential difference across such a liquid, strong electric fields may arise around those micropores, which would lower the molecular dissociation threshold and, in such a way, affect chemical reaction conditions. Cavitation-induced microheterogeneities in a liquid can also weaken the breakdown conditions and promote an electric discharge on the liquid surface. On the other hand, the ultrasonic stimulation of chemical reactions, which are atypical under normal conditions, in a liquid can affect the composition of the medium, in which plasma is formed, and thus change the plasma properties.

Therefore, plasma-liquid systems based of the DGCLW with the additional excitation of an ultrasonic field are rather interesting for the study and development. This work aimed at experimental researches of the properties of just such systems.

## 2. Experimental Installation and Technique

The experimental installation for studying a plasma-liquid system, which is based on the gas discharge immersed into a liquid with an ultrasonic field, is depicted in Fig. 1. It consists of quartz cylinder (*1*) hermetically sealed at its end faces with metal flanges. Cylindrical metal electrode (*3*) in quartz insulator (*4*) is introduced

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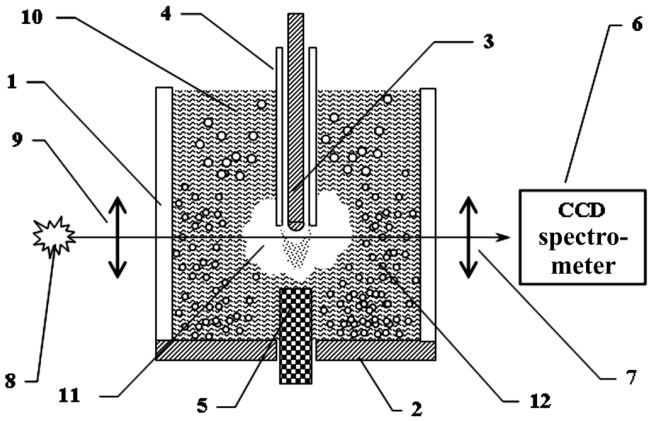


Fig. 1. Scheme of the experimental installation for studying a plasma-liquid system based on the gas discharge immersed in a liquid with ultrasonic field

through an aperture in the upper flange into liquid layer (*10*), being directed along the geometrical axis of the cylinder. In order to create gas channel (*11*) in the liquid, a gas is supplied through the space between the external lateral surface of the electrode and the internal lateral surface of the cylindrical quartz insulator. Lower metal flange (*2*) is immersed in the liquid and serves for the second electrode, which the discharge potential is applied to. In this work, we considered a regime, when the negative potential is applied to the liquid. Hence, the gas discharge breakdown takes place between metal electrode (*3*) and the surface of the liquid surrounding the dynamic gas channel. Magnetostriction ultrasonic projector (*5*) is introduced through a rubber seal in the central aperture of flange (*2*) into the liquid layer. The resonance frequency of projector was 17 kHz. The ultrasonic projector was excited with the help of a laboratory generator of sound frequency and a 100-W amplifier. Distilled water was a liquid to study. In the course of researches, the discharge current was maintained at a constant level of about 300 mA. The electroconductance of distillate increased with the plasma exposure time. Therefore, the voltage between electrodes *2* and *3* changed within the interval 2.5–1.8 kV. The ultrasonic radiation intensity during experiments was maintained above the cavitation threshold. Air was used to create the gas channel.

The plasma parameters were studied taking advantage of the emission spectroscopy method. The optical axis of the emission spectrum registration system passed through the midpoint of the gap between electrode (*2*) and ultrasonic radiator (*5*). The plasma radiation passed along the optical axis of the system through the working liquid layer and, with the help of a quartz lens (*7*), was focused on entrance slit (*6*) of a spectrometer.

The registered emission spectra could be distorted by the influence of optical characteristics of the liquid, which vary in time owing to the discharge action. The most essential factor, which can affect the radiation distribution over wavelengths, is the temporal changes in the absorption spectra of a working liquid, which stem from the plasma-induced changes of the liquid chemical composition. In addition, in the course of a discharge, the liquid bulk became filled very quickly with bubbles of gaseous products, which appeared owing to the electrolytic decomposition of the liquid. The passage of radiation through such a turbid medium can also manifest itself in a modification of the shape of emission spectra.

Since those factors influence the radiation independently, the role of each of them can be examined separately. It is known [10] that the intensity $I_r$ of radiation passed through a turbid liquid can be described by the equation

$$I_r = I_0 \times 10^{-k\frac{Cbd^3}{d^4+\alpha\lambda^4}}, \qquad (1)$$

where $I_0$ is the initial radiation intensity, when the radiation enters the liquid; $C$ the concentration of scattering centers; $b$ the thickness of the liquid layer; $d$ the average diameter of scattering centers; $\lambda$ the radiation wavelength; and $k$ and $\alpha$ are constants. For the system described in this paper, dispersion centers are gas bubbles. Since their characteristic dimensions are much larger than the wavelength, the term $\alpha\lambda^4$ can be neglected. Therefore, the presence of gas bubbles in the liquid layer reveals itself in the emission spectrum as an attenuation coefficient independent of the wavelength.

To take the influence of a changing chemical composition of the liquid into account, we registered absorption spectra. For this purpose, the radiation produced by standard light source (*8*) was directed, with the help of quartz lens (*9*), along the optical axis through the working liquid layer and, with the help of another quartz lens (*7*), focused on the entrance slit of spectrometer (*6*). A signal from the standard source was registered before the discharge ignition and immediately after its turning off. The emission spectrum was registered in a fixed time interval after the discharge ignition. During the processing of a liquid with plasma and the registration of emission spectra, the standard light source was switched off. A series of experiments were carried out for various time intervals of the plasma–liquid interaction in the regimes with and without an ultrasonic field in the liquid.

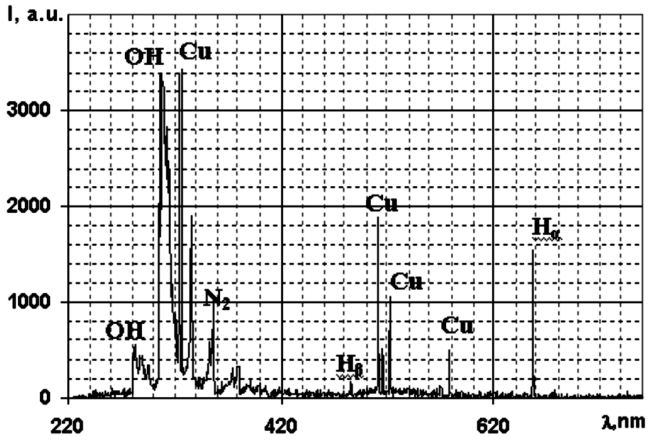


Fig. 2. Typical emission spectrum produced by the plasma of a gas discharge immersed in a liquid

## 3. Experimental Results

As a result of our experiments, we obtained the emission spectra of DGCLW plasma, a typical example of which is shown in Fig. 2. The spectra contain the lines of copper (a material the electrode was made of), the atomic hydrogen lines $H_\alpha$ and $H_\beta$ of the Balmer series, the molecular bands of the second positive system of nitrogen ($C^3\Pi$-$B^3\Pi$), and the molecular bands of hydroxyl ($A^2\Sigma$-$X^2\Pi$). The ratios between various components in the emission spectrum were found to change with the exposure time of the working liquid to a discharge.

Spectrophotometry methods were used to obtain absorption spectra of the liquid subjected to the action of DGCLW plasma. A typical example of these spectra is shown in Fig. 3,*a*. It was found that the absorption spectra are one-component. They correspond to the absorption band of nitrous acid ions and do not change their shape, irrespective of whether the ultrasonic field is present in the liquid or not. As the exposure time increased, a monotonous growth of the registered absorption peak was observed. In Fig. 3,*b*, the dependences of the absorption factor at a wavelength of 380.671 nm on the time of the distillate exposure to DGCLW are depicted. They were obtained in both regimes (presence or absence) of an ultrasonic field in the liquid. The curves testify that the concentration of nitrous acid that was synthesized under the discharge action is in a direct proportion to the time of the exposure with plasma. It is also clear that the synthesis of nitrous acid is more effective in the presence of ultrasound.

The dependences obtained for the absorption factors were used to correct the emission spectra with regard for the unstable chemical composition of a working liquid.

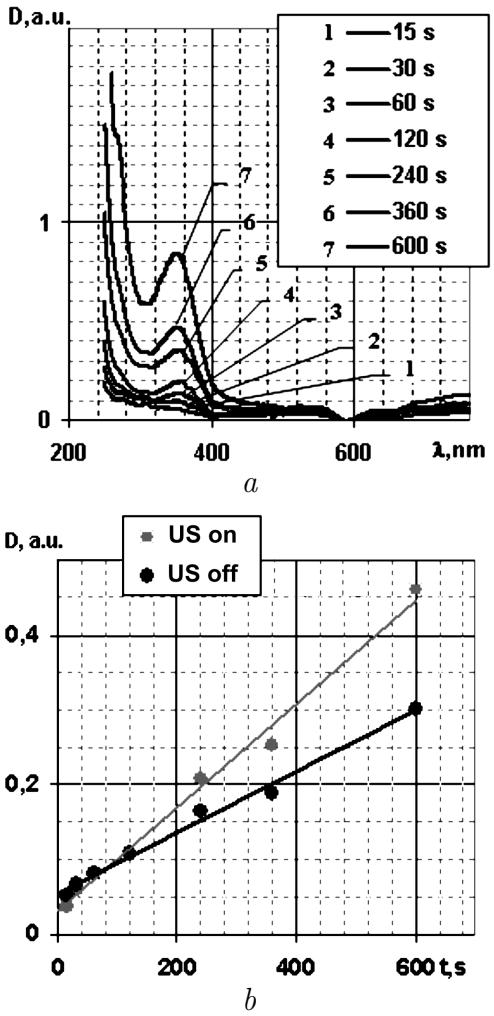


Fig. 3. (*a*) Emission spectra of distilled water treated with plasma for various treatment times. (*b*) Dependences of the absorption factor at a wavelength of 380.671 nm on the treatment time

In addition, in order to correct the influence of the light scattering at gas bubbles on the emission spectra, the intensities of all spectral components under investigation were normalized by the intensity of a line of atomic copper at a wavelength of 578.913 nm.

By analyzing the emission spectra, we obtained the dependences on the exposure time for the relative intensities of hydroxyl (Fig. 4) and nitrogen second positive system (Fig. 5) bands, as well as the atomic hydrogen line $H_\alpha$ (Fig. 6,*a*). We also used the ratio of the intensities of hydrogen lines $H_\alpha$ and $H_\beta$ to estimate the population temperature $T_e$ of hydrogen electron states. Fig. 6,*b* demonstrates the dependence of this temperature on the exposure time.

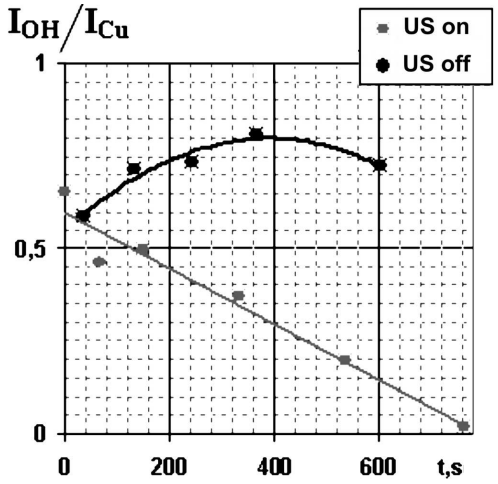


Fig. 4. Dependences of the relative intensity of the hydroxyl molecular band in the emission spectra of DGCLW plasma on the distilled water treatment time

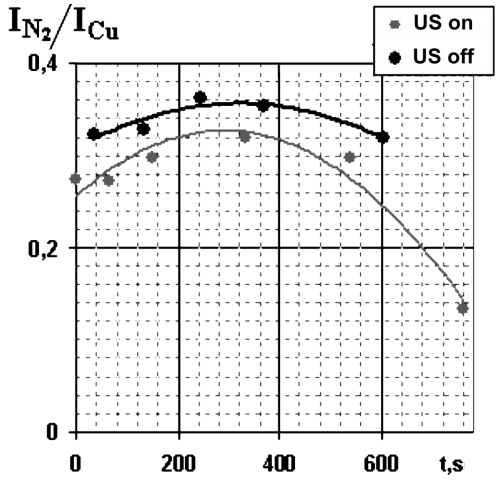


Fig. 5. Dependences of the relative intensity of molecular band $2^+$ of the nitrogen system in the emission spectra of DGCLW plasma on the distilled water treatment time

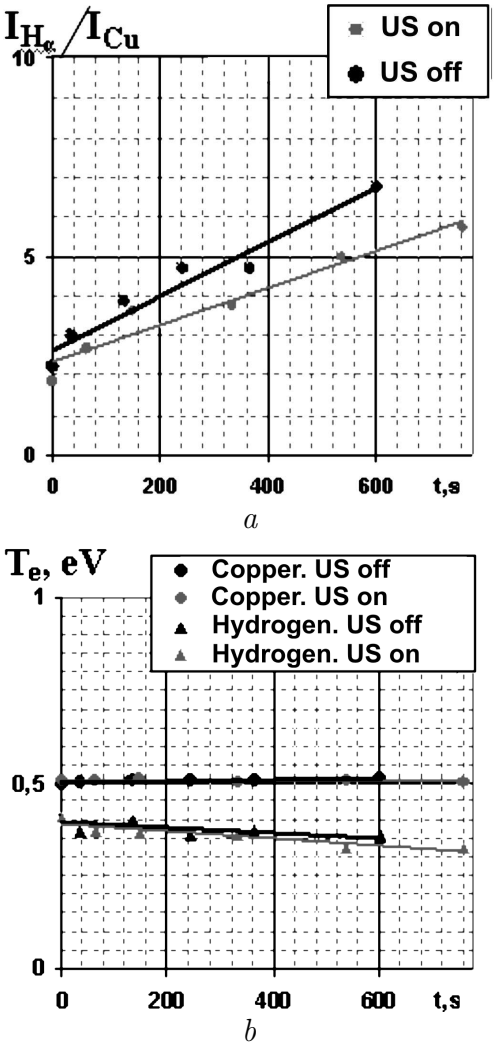


Fig. 6. Dependences of the ratio between the intensities of spectral lines of hydrogen $H_\alpha$ and copper ($a$) and the population temperatures $T_e$ for electron states of hydrogen and copper ($b$) in the emission spectra of DGCLW plasma on the distilled water treatment time

Ultrasound was found to have different effects on the behavior of different components in the emission spectra of DGCLW plasma. In particular, one can see from Fig. 4 that the relative intensity of radiation by hydroxyl in the absence of ultrasound grows nonmonotonously with the exposure time, but decreases rather quickly under the ultrasound action. The relative radiation intensity of nitrogen (Fig. 5) weakly depends on the exposure time in the absence of ultrasound, but nonmonotonously changes in the presence of ultrasound, reaching a maximum after the 360-s exposure. The relative intensity of line $H_\alpha$ grows linearly with the exposure time in both regimes (Fig. 6,$a$). However, ultrasound reduces the rate of such a growth by a factor of one and a half.

The growth of the relative intensity of radiation by hydrogen is not accompanied by the growth of the population temperature $T_e$ for its electron states (Fig. 6,$b$). Ultrasound practically does not influence the $T_e$-behavior.

It is important to note that every component in the emission spectra demonstrates a little lower relative intensity of radiation in the presence of ultrasound. It can be explained by the fact that, at frequencies of about 17 kHz, ultrasound can propagate in air. Moreover, a stationary contact between DGCLW plasma and the liq-

uid surface gives rise to the transformation of the sharp phase interface into an intermediate layer, the properties of which can promote the penetration of ultrasound into the gas phase. The presence of the ultrasonic field in plasma can increase the probability of the excited atomic state quenching by means of the collision mechanism, which would bring about a reduction of the average number of radiation events per unit time. Hence, a small reduction of the total intensity of the radiation by DGCLW plasma under the action of ultrasound can be associated with the penetration of ultrasonic field into the plasma phase.

## 4. Conclusions

As a result of our researches, we may assert the following.

The ultrasonic field in the liquid phase of a plasma-liquid system based on a discharge in the gas channel with a liquid wall enhances the efficiency of the nitrous acid production in a working liquid by a factor of approximately 1.5.

The ultrasonic field in the working liquid differently affects the evolution of composition components in the radiation spectrum of DGCLW plasma. In particular, the ratio between radiation fractions of hydroxyl and nitrogen molecules decreases substantially.

In plasma-liquid systems based on a discharge in the gas channel with a liquid wall, a linear growth of the relative intensity of atomic hydrogen lines with the plasma–distilled water interaction time is observed. The ultrasonic field makes the rate of this growth about 1.5 times slower.

The ultrasonic field in the liquid phase of the plasma-liquid system based on a discharge in the gas channel with a liquid wall practically does not affect the population temperature of electron states in hydrogen.

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ВПЛИВ УЛЬТРАЗВУКОВОГО ВИПРОМІНЮВАННЯ НА ВЛАСТИВОСТІ ПЛАЗМИ ГАЗОВОГО РОЗРЯДУ, ЗАНУРЕНОГО В РІДИНУ

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Р е з ю м е

Роботу присвячено експериментальним дослідженням властивостей плазмово-рідинних систем на базі електричного розряду в газовому каналі з рідкою стінкою з додатковим збудженням ультразвукового поля в рідині. Показано, що наявність ультразвукового поля в рідинній фазі таких систем підвищує ефективність напрацювання азотистої кислоти та впливає на еволюцію компонентного складу випромінювання плазми.