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FEATURES OF WATER TREATMENT USING ELECTROPHYSICAL AND PLASMA-CHEMICAL EFFECTS

M.I. Boiko[1], A.V. Makogon[2]
[1]National Technical University "Kharkiv Polytechnic Institute", Kharkiv, Ukraine;
[2]Institute of Electrophysics and Radiation Technologies NAS of Ukraine, Kharkiv, Ukraine
E-mail: qnaboyg@gmail.com, artemmak1991@gmail.com

Accumulated experimental results on water treatment using nanosecond discharges in air bubbles confirm the practical effectiveness of this approach, while numerical modelling of a negative streamer in the «air bubble–water» system reveals the physics of its formation. Computer simulation has demonstrated the distributions of electric potential and electric field strength, the evolution of space charge, the accumulation of surface charge at the phase interface (~0.017 C/m²), the characteristics of current flow in the reactor (electron, displacement, and ion currents), and has provided a detailed analysis of the role of the multiphase medium in the formation of such streamers. Water treatment technology based on nanosecond discharges in air bubbles represents a promising foundation for industrial installations and opens broad opportunities for further theoretical and experimental research.

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INTRODUCTION

Ensuring effective disinfection of food products, as well as water decontamination and purification, often of high electrical conductivity, in various branches of industry, the energy sector, and marine transport is an extremely relevant challenge today.

It is known that under the operating conditions of nuclear power plants (NPPs), a significant volume of water is used, including water from recirculating cooling systems and storage reservoirs, in which microorganisms, organic matter, and biofilms may actively develop. Such water can pose a biological hazard and therefore requires appropriate treatment [1–4]. Traditional disinfection methods (chemical and non-chemical) provide insufficiently effective treatment and have their own drawbacks. At present, physicochemical methods based on electrical discharges in liquid and air-water media are being actively investigated for the disinfection of such water [5, 6].

Methods of ozonation and ultraviolet treatment have also been well studied and have long proven effective for large-scale industrial application [7–10]. Ultraviolet radiation causes damage to the DNA and RNA of microorganisms, while ozone, owing to its high oxidative capacity, decomposes complex organic contaminants and oxidizes bacterial cell membranes, leading to their destruction. However, ozone generators utilize only part of the potential of electrical discharges, and ultraviolet treatment, although simple to operate and free of chemical additives, shows low efficiency in water with increased turbidity or color. Particles and impurities in the water absorb and scatter the radiation, hindering its penetration. In addition, ultraviolet treatment does not provide prolonged protection; therefore, after treatment, without additional disinfection, there is a high probability of pathogen regrowth.

One of the most promising approaches is the application of high-voltage pulsed discharges. High-voltage discharges produce a complex effect through a strong electric field and accompanying ultraviolet radiation, while also generating reactive species with high oxidative potential, including ozone, which makes it possible to significantly enhance the efficiency of water disinfection and purification and improve the energy efficiency of the treatment process. To maximize the effect of the most efficient discharge factors, it is advisable to generate the discharge in gas bubbles directly within the water being treated [11, 12], since the lifetime of species such as OH radicals, with an oxidation potential of 2.8 V, and atomic oxygen O, with an oxidation potential of 2.42 V, is less than one millisecond.

This approach is innovative and has strong potential for the effective treatment of wastewater, ballast water in marine transport, coke-plant water, and similar media. In designing industrial reactors and installations, it is necessary to take into account the specific features of high-voltage pulsed discharge formation under such load conditions, as well as to provide additional means of current protection and protection of the electrodes against the corrosive effects of the aggressive medium.

The aim of this work is to theoretically investigate the specific features of the formation of electrophysical and plasma-chemical factors in a reactor during liquid treatment by discharges with nanosecond rise times in air bubbles.

EXPERIMENTAL RESULTS

The authors developed and tested a method for the disinfection and purification of liquids using pulsed electrical discharges in air bubbles within the liquid [13–15]. At the present stage, an experimental prototype has been created to implement this approach, featuring a pipeline reactor for in-flow water treatment with a nominal pulse power of 3 MW. To monitor pulse parameters (current, voltage, and frequency), a capacitive voltage divider with a division ratio of K_d=7653, a shunt resistor with resistance R_s =2.5 Ω, and a RIGOL DS1102E digital oscilloscope with a bandwidth of 100 MHz were used. Under nanosecond discharges, the voltage amplitude across the load in the form of an air bubble in water reaches 35 kV, while the current amplitude reaches 100 A, at a pulse repetition frequency

of $f \approx 2200$ Hz, a pulse duration of approximately 60 ns, and rise times up to 10 ns.

Fig. 1 shows photographs of the typical electrode configurations used during water treatment.



Fig.1. Typical electrode configurations used during water treatment

In Fig. 1, the high-voltage electrodes are designed in the form of a «brush», that is, a bundle of 6–10 central strands of a multicore copper cable with a diameter of D = 0.1…1.5 mm. Such a needle-like shape sharply increases the local electric field strength and facilitates the initiation of ionization processes. The tip of the «brush» is mounted in special holders, which makes it possible to quickly replace the tip and supply gas directly to the high-voltage electrode region. The low-voltage electrodes are usually made in the form of a copper plate measuring 30×200×1 mm, a metal ring (not shown in the photograph), or a cylindrical copper electrode with an opening, designed for installation in a flow-through pipeline reactor. The distance between the high-voltage and low-voltage electrodes can be smoothly adjusted within the range of 3…100 mm, making it possible to adapt the discharge to the specific electrical conductivity of the liquid and the treatment mode. Gas of any required composition can be supplied to the high-voltage electrode region, including atmospheric air, oxygen, inert gases (Ar, Ne), or their mixtures (such as air–ozone mixtures). The system also allows bubbling of the water being treated. So far, only atmospheric air has been used in the experiments, supplied from a compressor through a system of adjustable hoses.

Photographs of such discharges are presented in Fig. 2.



Fig. 2. Photographs of nanosecond discharges in air bubbles during water treatment

In Fig. 2, an air bubble in water with a diameter of 15…20 mm serves as a localized volume for plasma formation. The photographs are integral images showing the combined effect of different ionization intensities and streamer development in different regions; violet, pink-orange, and yellow-orange emission, characteristic of different plasma phases or zones, is superimposed within a single frame. A detailed analysis of the temporal dynamics of plasma structure development requires expensive high-speed imaging of the discharges with high temporal resolution, as well as spectroscopic studies of the discharge emission, which would make it possible to identify the dominant plasma-chemical mechanisms.

The air bubbles in which the discharges are generated rise through the water column, simultaneously saturating the water with strong oxidants and promoting additional mixing of the treated medium. Recent experimental studies by other authors on plasma–liquid interaction demonstrate that streamer impacts on the water surface can generate upward flows in weakly conductive treated media, whereas in solutions with increased conductivity, electrohydrodynamic effects and grounding of the solution produce downward jets, ensuring deeper penetration of active components to a depth of 1…3 cm at a velocity of up to 4 mm/s [16].

Our previous studies on the treatment of water used in coke production, as well as surface water, showed that water can be effectively purified by means of discharges in gas bubbles within it. In particular, the concentrations of thiocyanates were reduced by 94% and phenols by 60.3% (according to data from the laboratory of the Kharkiv Coke Plant). River water became clearer, its biochemical oxygen demand decreased, and undesirable odors were eliminated. The possibility of removing ammonia from water containing ammonium nitrate, NH_4NO_3, was also demonstrated. A reduction of 37% in ammonia (NH_3) concentration was achieved. In addition, 100% disinfection of E. coli bacteria in model water was obtained from an initial concentration of 10^6 CFU/cm^3 (according to data from the Municipal Enterprise «Sanepidservice», Kharkiv, CFU – colony-forming units).

When water has a high level of contamination, the use of nanosecond discharges directly in air bubbles within the water becomes especially promising, because they create a powerful combination of factors that makes it possible to effectively decompose a wide range of toxic impurities, reduce the concentrations of organic substances and ammonia, and completely disinfect the water.

DESCRIPTION OF THE COMPUTATIONAL MODEL

For numerical simulation, the computer model of negative streamer development presented in [17] was used.

Fig. 3 shows the graphical model of the reactor with the interelectrode gap and electrode configuration corresponding to Fig. 2.

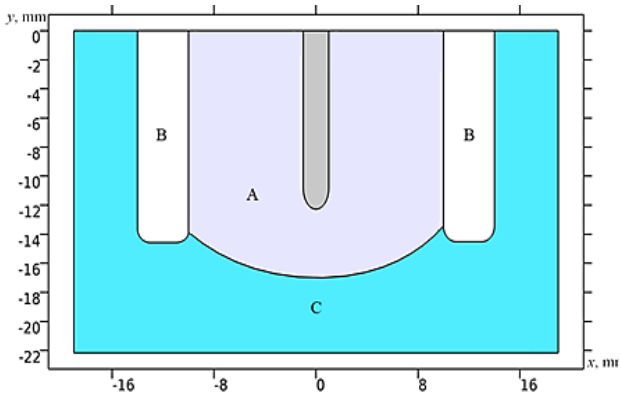


Fig. 3. Graphical model of the reactor

Region A is the air bubble, where plasma-chemical processes take place, modeled as an ideal gas, C is the treated liquid with a dielectric permittivity of 80, and B is the insulating housing with a relative dielectric permittivity of 2.3. The temperature of the gas, liquid, and housing was T = 300 K, and the pressure was one atmosphere.

The initial conditions in this study were as follows: the computational domain of the reactor was discretized using an unstructured triangular mesh with a characteristic element size of 0.015…0.2 mm, with additional mesh refinement in the high-voltage electrode region and inside the air bubble. Numerical simulation of plasma processes in the reactor was carried out over the time interval from 0 to 1.5 ns with a time step of $1 \cdot 10^{-4}$ ns, which made it possible to reproduce the nanosecond voltage rise front up to its peak amplitude (34 kV). At the initial moment of time, the concentration of positive ions n_i (N_2+, O_2+) was $3 \cdot 10^6$ cm^{-3}, the electron concentration n_e was $1 \cdot 10^6$ cm^{-3}, the concentration of negative ions n_i (N_2−, O−) was $2 \cdot 10^6$ cm^{-3}, and the initial electron energy was ε = 0.026 eV. The initial gas mixture in the bubble contained 77 % molecular nitrogen (N_2), 20 % molecular oxygen (O_2), and the remaining 3 % consisted of other plasma components.

SIMULATION RESULTS AND DISCUSSION

Fig. 4 shows the distribution of electric potential and electric field strength during streamer formation in the air bubble.

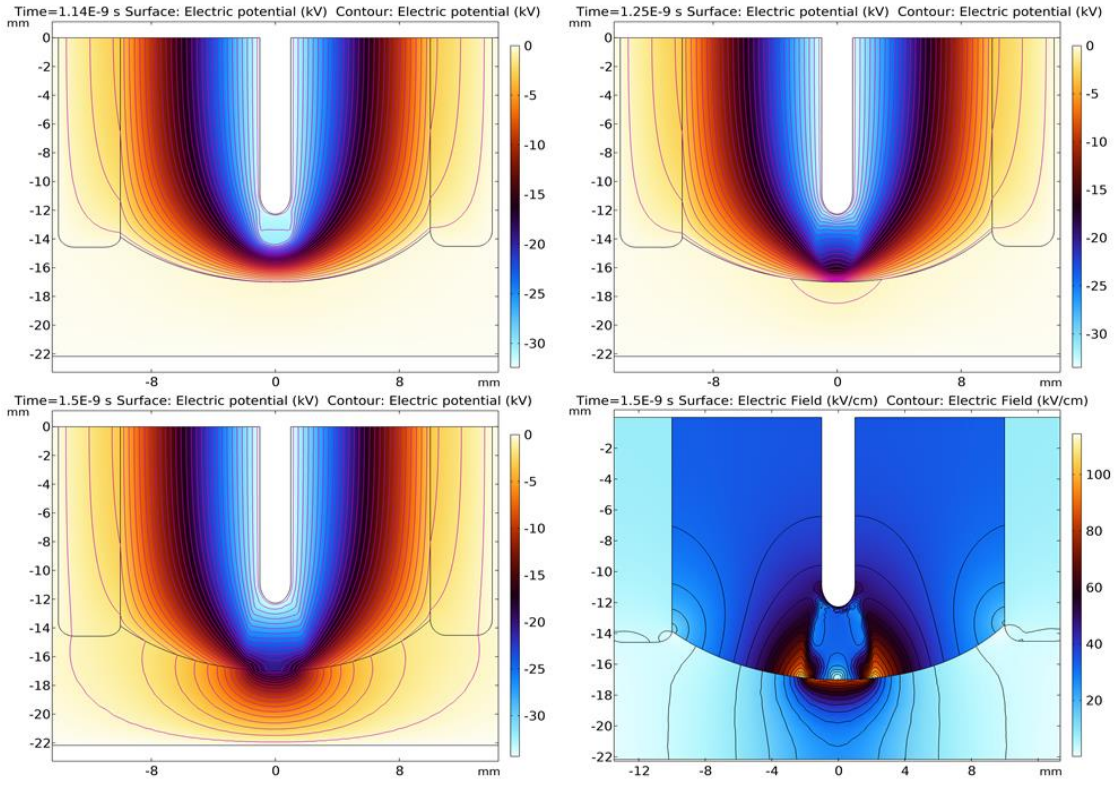


Fig 4. Distribution of electric potential and electric field strength

Fig. 4 considers the case where a voltage pulse with a rise time of 1.5 ns is applied to a 10 mm long "rod–plane at zero potential" discharge gap. Of these 10, 5 mm from the high-voltage rod is occupied by an air layer, while the next 5 mm up to the plane is occupied by a water layer. At the moment the voltage begins to rise, an electric field arises throughout the entire two-layer discharge gap of 10 mm length, with the field strength equal to the potential gradient. The electric field appears simultaneously in the water layer and in the air layer (inside the air bubble) as soon as the potential difference is established across the two-layer discharge gap. At the moment when the potential difference appears across the two-layer discharge gap, plasma in the gas layer (the air bubble) does not yet exist. Plasma in the form of a plasma structure – a streamer – appears in the air bubble later,

namely after the potential on the rod (negative in our case) reaches a certain value sufficiently high in magnitude. In this study, the indicated layers – the gas layer (air bubble) and the water layer – are considered as two series-connected capacitors with lumped parameters. Under these conditions, when the potential on the metal rod reaches a certain value, a plasma structure – a streamer – arises in the gas bubble and propagates toward the "gas bubble–water" interface. When this plasma structure, i.e., the streamer with sufficiently high active electrical conductivity, reaches this interface, most of the total potential difference across the entire two-layer gap is applied to the water layer. Therefore, the electric field strength in the water layer near the moment of contact increases sharply, taking into account that the field is also strongly nonuniform. It should be noted that the final distributions of electric potential and electric field strength in the reactor at the time instant of 1.5 ns reproduce the pattern established in the previous work [18], even when the initial concentrations of the main gas-phase components are varied.

Fig. 5 presents the calculated evolution of space charge during streamer formation.

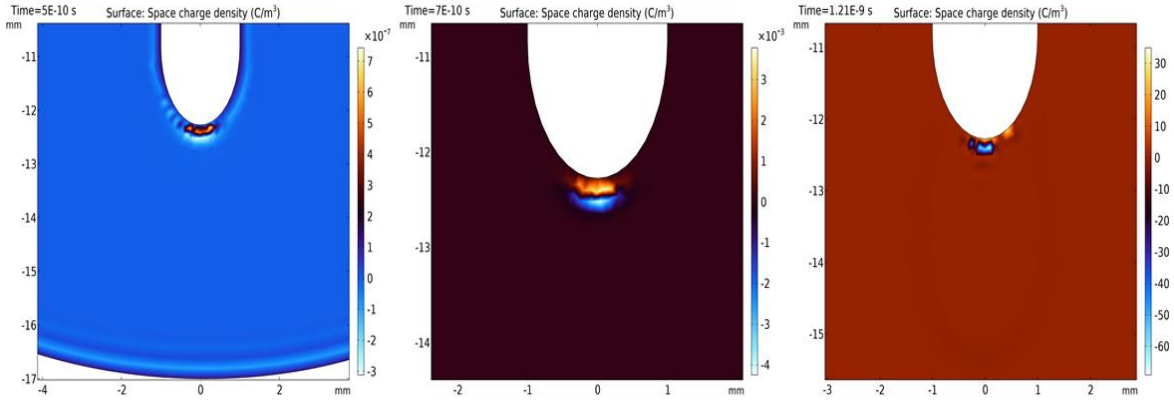


Fig. 5. Evolution of space charge during streamer formation

The presented numerical results (see Fig. 5) show the evolution of space charge during the formation of a negative streamer. The figures demonstrate that, in the course of streamer development, the quasineutrality of the plasma is disturbed. According to the work of Yu.P. Raizer [19], the initial state of the gas prior to the formation of streamer structures is characterized by a low degree of ionization and a volume charge density close to zero. At the moment a high-voltage pulse is applied, a strong electric field arises, accelerating free electrons to high energies and thereby enabling impact ionization of gas molecules. As a result, an avalanche-like increase in the concentration of charged particles (electrons and positive ions) occurs, and a space-charge region is formed. The magnitude of the space charge increases over time in a localized region near the electrode and the front of the negative streamer, where a strong nonuniformity in electron and ion concentrations develops. This mechanism sustains the self-propagation of the streamer, which manifests itself as a highly conductive plasma channel. One can also observe the formation of several space-charge layers, in which positively and negatively charged regions clearly alternate. The appearance of such structural features can be explained by differences in the physical properties of electrons and ions. In the calculations, electrons are characterized by a high temperature (tens of eV) and extremely high mobility, whereas the heavy ions in the model have a temperature of 300 K, corresponding to the gas temperature. At the same time, quasineutrality is preserved in the rest of the plasma volume.

Fig. 6 presents the results of numerical simulation of the evolution of surface charge at the "air bubble (plasma)–water" interface during the formation of a negative streamer.

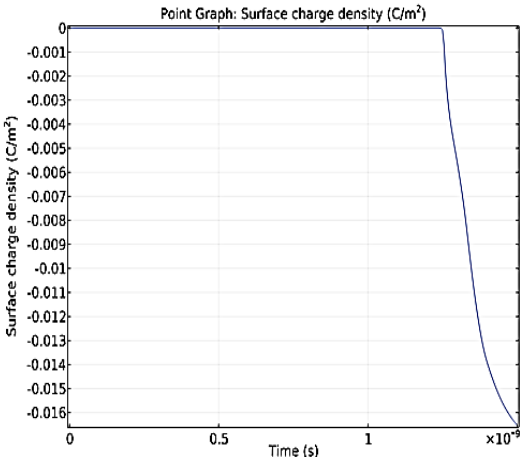
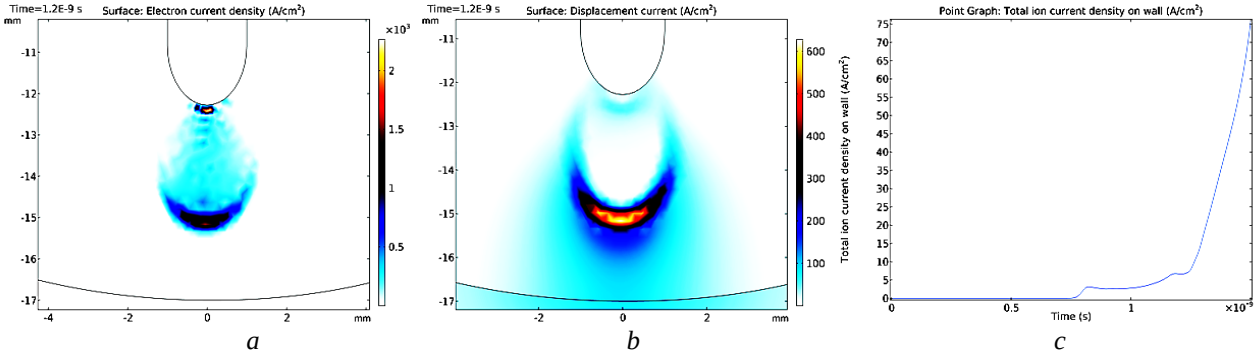


Fig. 6. Surface charge at the "air bubble (plasma)–water" interface

As can be seen from the graph in Fig. 6, at the initial stage (up to approximately 1.25 ns), no surface charge accumulates and its value remains close to zero, because the front of the negative streamer has not yet reached the water surface and plasma processes occur mainly within the internal volume of the air bubble. After the streamer front reaches the water surface (at approximately 1.25 ns), the magnitude of the negative surface charge increases sharply, reaching a maximum value of -0.017 C/m^2. Water, which has high dielectric strength and high permittivity, acts as a dielectric barrier that prevents further penetration of the streamer channel, leading to charge accumulation at the phase interface. Such accumulation of negative surface charge creates a localized electric field that significantly affects the subsequent development of the discharge. The local field enhancement at the "gas (plasma)–water" interface

promotes the formation of sliding discharges along this boundary, contributing to the emergence of additional zones of impact ionization. The accumulated surface charge may also play an important role in the activation of the aqueous medium through the generation of reactive species, including OH radicals.

Fig. 7 shows the distributions of current densities flowing in the reactor during streamer formation.



a *b* *c*

Fig. 7. Distribution of current densities in the reactor during streamer formation: a – electron current density; b – displacement current density; c – ion current densities at the surface of the high-voltage electrode

At the time instant of 1.2 ns (see Fig. 7,a,b), the mechanism of current flow in the reactor is shown. Directly within the streamer, the current is determined by electron motion, whereas below the streamer and in the water, displacement currents predominate. Fig. 7,c presents the calculated values of ion current density at the surface of the metal rod. Inside the streamer plasma channel, which by this moment has extended by approximately 3 mm from the metal electrode, a high concentration of electrons and positive ions is observed. Below the streamer and in the water, the concentration of free charge carriers remains insignificant, and the current is mainly determined by the variation of the electric field, that is, by the displacement current. The ion current at the rod surface arises at a later stage of the process: up to 0.75 ns, its density near the tip is practically zero, after which the first drifting ions appear in the streamer tail, and at 1.2 ns its value increases sharply.

The accumulated experimental results confirm the promise of this technology and open up opportunities for its scaling and industrial implementation, while numerical modeling of the negative streamer in the "air bubble (plasma) – water" system reveals the physics of its formation, demonstrates the necessity of space charge for streamer initiation, and shows the characteristic features of the development of such discharges in multiphase media.

CONCLUSIONS

1. By means of computer simulation, the formation of a negative streamer in an air bubble in water under the action of a nanosecond pulse with a 1.5 ns rise time and a voltage amplitude of 34 kV was investigated. The influence and role of the "gas (plasma)–liquid" interfacial boundary were analyzed. At this boundary, an intense localized electric field is formed, negative surface charge accumulates after the streamer front reaches the water interface, and conditions are created for the occurrence of sliding discharges along the water surface.

2. It was shown that streamer development is accompanied by a breakdown of plasma quasineutrality and the formation of localized space-charge regions, which determine the electrophysical characteristics of the discharge and promote its further propagation. The features of current distribution in the reactor were analyzed. Within the streamer plasma channel, electron conductivity predominates, whereas below the streamer and in the water, the decisive role is played by the displacement current caused by the capacitive nature of the multilayer structure "high-voltage electrode – streamer – air bubble – liquid – low-voltage electrode".

3. The simulation results, as well as the presented accumulated experimental data on liquid treatment by means of high-voltage nanosecond discharges in air bubbles, confirm the prospects of industrial-scale liquid disinfection and purification, wastewater treatment and inactivation at energy facilities, ballast water treatment in marine transport, final purification of drinking water, and water disinfection in swimming pools and other facilities with increased sanitary and epidemiological requirements, and demonstrate a strong need for further research.

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ОСОБЛИВОСТІ ОБРОБКИ ВОДИ ЗА ДОПОМОГОЮ ЕЛЕКТРОФІЗИЧНИХ ТА ПЛАЗМОХІМІЧНИХ ВПЛИВІВ

М.І. Бойко, А.В. Макогон

Накопичені експериментальні результати обробки води за допомогою наносекундних розрядів у повітряних бульках підтверджують прикладну ефективність підходу, а чисельне моделювання негативного стримера в системі «повітряна булька – вода» розкриває фізику його формування. Шляхом комп'ютерного моделювання показано розподіли потенціалу та напруженості електричного поля, зміну просторового заряду, накопичення поверхневого заряду на границі розділу фаз (~0,017 Кл/м²), особливості протікання струмів у реакторі (електронного, зміщення, іонного) та детально проаналізовано роль багатофазного середовища при формуванні таких стримерів. Технологія обробки води на основі наносекундних розрядів у повітряних бульках є перспективною основою для промислових установок і відкриває широкі можливості для подальших теоретичних та експериментальних досліджень.