

Review

# Application of Water Treated with Low-Temperature Low-Pressure Glow Plasma (LPGP) in Various Industries

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**Abstract:** Plasma processing is now a key technology across the world, and nonthermal low-temperature plasmas are being increasingly used. This situation can be explained by a rapidly growing interest in the optimization of existing methods, as well as the development of new ones. Over the last few years, the production of plasma-treated water (PTW) by low-temperature low-pressure glow plasma (LPGP) under an atmosphere of various gases has been increasingly gaining in popularity. Research has been conducted on producing plasma-treated water in the presence of air, nitrogen, ammonia, carbon dioxide, and methane. All the obtained results show that the changed physicochemical properties of the water depend on the type of gas used and the duration of the plasma treatment. New research is emerging on the possibility of using this water in plant breeding, animal husbandry, cosmetology, medicine, and food. For the first time, plasma-treated water has also been tested for use in the brewing industry at the raw material preparation stage. The results obtained in all branches of science are very promising, contributing to the growing interest in plasma-treated water within the scientific community.

**Keywords:** water; macrostructure reconstruction; clathrate hydrates; glow plasma; plasma-treated water



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## 1. Introduction

Plasma is ionized matter with a gas-like aggregation state in which a large proportion of the particles are electrically charged [1]. In most cases, plasma is a poorly ionized gas with a low degree of ionization [2]. Plasmas are generated by thermal energy, electric field energy, radiation, or beams (lasers, UV photons, electrons, or protons) [3]. These are particle systems in the form of liquid or gaseous mixtures of free electrons and ions, which may also contain neutral particles (atoms or molecules) of high average kinetic energy or solely charged plasma components [4]. Plasma affects matter by modifying it. Cold plasma is used to modify the surface of materials, leading to simple topographic changes forming surface and coating chemistry, in this case, in water. During the process of excitation/ionization of gaseous products, plasma energy molecules break chemical bonds by colliding with the material surface, generating free radicals on the surface. They are subject to additional reactions that depend on the type of plasma gas used. This process produces surfaces that have different properties in relation to the mass of the material. When controlling the plasma treatment process, parameters such as treatment time, power and type of gas, pressure, and frequency should be taken into account [5]. Plasma, depending on the pressure and temperatures at which it occurs, can be divided into high- and low-temperature forms [6]. Under terrestrial conditions, low-temperature plasma can be obtained. This plasma is not fully ionized and contains some neutral particles [7]. Low-temperature plasma can be divided into thermal (equilibrium) and nonthermal (nonequilibrium) forms [8].

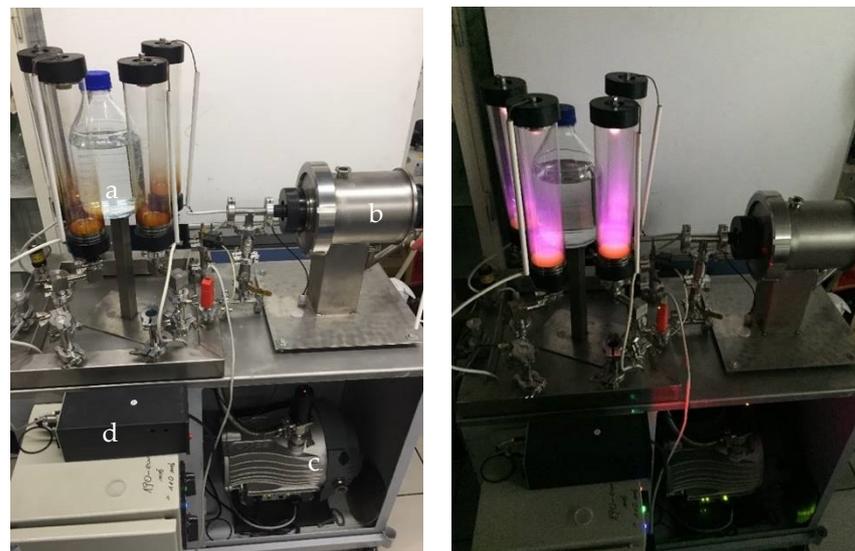
The processing of nonthermal plasma (NTP) under atmospheric conditions has recently gained a lot of interest among researchers. This is especially true when it comes to

the use of NTP in food production [9]. NTP has bactericidal properties and can also be used for the removal of pesticide residues [10,11]. In the food industry, the NTP is mainly used in the minimal processing preservation method, which reduces the risk of microbiological contamination and spoilage [6,12].

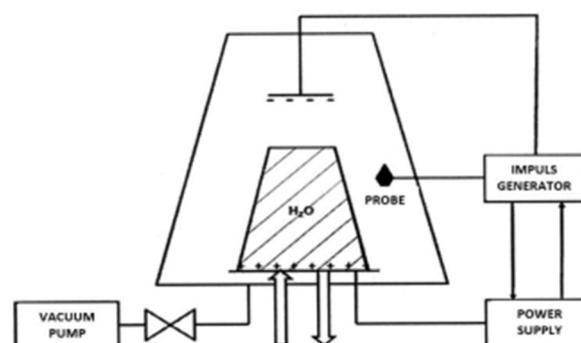
Under the influence of high voltage, the conductivity of water (polar liquid) is changing [13], which makes the application of NTP to a wet surface or a liquid substrate a more complicated process [14,15]. The presence of water molecules during NTP treatment affects the chemical properties of reactive species and their interaction with target microorganisms. The properties of NTP also depend on the type of carrier gas used during the plasma generation [16]. During the production of NTP, the pH value changes depending on the time and volume of the solution. An acidic environment in the presence of nitrates enhances the antimicrobial properties of the solution [17].

## 2. Treating Water with Low-Temperature, Low-Pressure Glow Plasma (LPGP)

Until now, there has been no known device for treating liquid substances with cold plasma at room temperature. For this purpose, in 2014, Polish scientists patented a device for the production of low-temperature, low-pressure, low-frequency glow plasma (LPGP) shown in Figures 1 and 2 [18,19]. This device was initially used for water treatment.



**Figure 1.** Plasmothrone (without cover) used in this study. (a) Lamps, (b) pulse generator, (c) vacuum pump, and (d) power supply [19].



**Figure 2.** Scheme of the glow plasma (GP) generator and the mode of treating water with it [18].

Plasma-treated water (PTW) is water that has been exposed to low-temperature, low-pressure, low-frequency glow plasma. This water is obtained under laboratory conditions by placing a glass bottle (e.g., a Pyrex bottle) with a capacity of up to 2000 mL into the

reactor chamber near the plasma lamp. In the reactor chamber, the water is irradiated for a specified period of time (from 5 to 90 min). The lamp generates plasma at 38 °C under a pressure of  $5 \times 10^{-3}$  mbar, a voltage of 600 V at 50 mA, and a frequency of 10 kHz. For this purpose, distilled, deionized, and tap water of various mineralization levels and hardness levels have been used. The produced water is stored at ambient temperature in closed Teflon containers with a capacity of 2000 mL [19,20].

During the entire process, large systems—gigaclusters—of water break down. This treatment leads to water with different physicochemical properties being obtained, e.g., increasing the electromagnetic conductivity permittivity (epsilon), or reducing the surface tension. These changes affect, among other properties, the absorption of water by living organisms, allowing for greater absorption [21]. The process that takes place in water after the appropriate treatment with cold plasma is related to the effect of water declustering. The physicochemical properties of plasma-treated water depend on the treatment process, so when controlling the entire process, the following parameters should be taken into account: treatment time, plasma power and type, gas type, pressure, and frequency [5].

In order to check whether water exposed to low-temperature, low-pressure glow plasma (LPGP) has changed physicochemical properties, a number of analyses are typically carried out. The obtained water samples are characterized by pH value, conductivity, surface tension, differential scanning calorimetry (DSC), water density, FTIR-ATR (Fourier-Transform Infrared Spectroscopy-Attenuated Total Reflectance) Spectra, ESR (Electron Spin Resonance) Spectra, UV/VIS (Ultraviolet/Visible) Absorption Spectra, and Raman Spectra. Thanks to these analyses, significant differences between the produced water and untreated water can be observed [22].

### 3. Structure and Physicochemical Properties of Plasma-Treated Water in the Presence of Various Gases

Until now, the plasma treatment of water has been carried out with the water in contact with air, but now new research has been carried out on the plasma treatment of water under atmospheres of nitrogen, ammonia, carbon dioxide, or methane. Initial research by Mystkowska et al. [22] on the influence of LPGP on deionized water showed that water exposed to low-temperature, low-pressure glow plasma had significantly changed physicochemical properties. The pH value, electrical conductivity, and surface tension changed in comparison to the control sample (not exposed to plasma). However, the origin of the observed effects has not been elucidated, and the structure of the PTW (Plasma-Treated Water) remained unknown. Białopiotrowicz et al. [23] presented the structure and selected physicochemical properties of water exposed to LPGP in air. They proved that the treatment of water with low-temperature, low-pressure glow plasma changes the macrostructure of the water to a degree that depends on the duration of the water's exposure to the glow plasma. The resulting water is an aqueous solution of clathrates with singlet oxygen molecules contained in them. The structure of the obtained clathrates also depends on the duration of the water's exposure to the glow plasma. It has also been proven that PTW is stable in long-term storage, survives cooking and contact with alkalis, but is unstable in acidic solutions. In studies conducted by Tomasiak [20], it was shown that water treated with glow plasma (GP) is characterized by a discoloration of its macrostructure, and the molecules of gas dissolved in this water are in an excited state. Chwastowski et al. [24] treated water with plasma in the presence of anaerobic nitrogen. The deionized water samples were treated from 5 to 90 min with a low-temperature, low-pressure glow plasma (GP). This operation made it possible to obtain nitrogen in various excited states. The oxygen and carbon dioxide that were already dissolved in the water were eliminated from the deionized water by clathrates that were generated with various nitrogen molecules in an excited state. This change resulted in a higher order of the water macrostructure, which was confirmed by the appropriate Raman spectra. Ciesielska, A. et al. [25], in another paper, extended the research to the operation of low-temperature, low-pressure glow plasma in the presence of methane. The obtained samples had completely

different physicochemical properties than the samples obtained for plasma-treated water in the presence of air, ammonia, or nitrogen. Removal of the air dissolved in the water by methane purging increased the number of water molecules vibrating asymmetrically. The resulting water clathrates did not contain methane molecules. The most significant changes were observed for water treated with plasma for 15 min. In another article by Ciesielska, A. [26], attention was focused on studying the structure and properties of water treated with low-temperature, low-pressure glow plasma in the presence of carbon dioxide. In this area of research as well, significant changes to the physicochemical properties of water were observed, compared to the control sample (not exposed to plasma). In the article devoted to the study of water exposed to low-temperature, low-pressure glow plasma in the presence of ammonia [27], it was observed that the formed water did not influence the formation of the boiling scale and corrosion, which contributes to the protection of water pipes. Thanks to this research, it was proven that not-commonly known chemical, physical, and physicochemical methods can be used to reduce the boiling scale and corrosion. In addition, water treated with low-temperature, low-pressure glow plasma in the presence of ammonia can serve as an alternative, environmentally friendly nitrogen fertilizer. In his subsequent research, Chwastowski [28] focused on the study of deionized and tap water with and without (control sample) an increased content of molecular oxygen (WST) subjected to low-temperature, low-pressure glow plasma (LPGP). The saturation of the water with oxygen before the plasma process turned out to be more advantageous compared to the saturated water after the treatment with the glow plasma. At this stage of the research, scientists concluded that tap water dissolves oxygen better than deionized water. The results obtained were also dependent on the duration of the treatment of the respective samples with plasma. In studies conducted by Ikawa et al. [29], it was shown that PTW had a strong bactericidal activity against *Escherichia coli*. This effect was enhanced under acidic conditions (peritonic acid), which changed the physical properties of the obtained water, increasing its bactericidal effect. Shaw et al. [30] highlighted the role of reactive nitrogen species (RNS) in the inactivation of bacteria. For this purpose, they developed a plasma system with increased sterilization efficiency. In the conducted research, it was noticed that the plasma generated in the presence of vapors, and especially  $N_2 + 0.5 \text{ wt}\% \text{ HNO}_3$ , was more effective at inactivating bacteria than PTW was. Bosch et al. [31] investigated the effectiveness of PTW in the treatment of a mealybug (*Planococcus citri*). The water was exposed to plasma for several minutes (1, 3, 5, and 10 min). After this time, a clear acidification of the water was determined, which resulted in a pH value of 1.8 after 10 min of treatment. Such a preparation of PTW led to a high mortality rate of approx. two thousand, 90% after a 24 h follow-up period.

#### 4. The Use of Low-Temperature, Low-Pressure Glow Plasma in Various Fields of Science

Water treatment with low-temperature, low pressure glow plasma (LPGP) is a promising technology in various fields of science. For several years, research has been conducted on the use of plasma-treated water, so-called nanowater, in animal husbandry and plant breeding. Murawski et al. [32] checked the effect of plasma-treated water on insemination in animal husbandry. Semen collected from animals before use is frozen with liquid nitrogen in an aqueous suspension. In this article, the researchers reported preserving the semen of rams. After thawing the samples, both in plasma-treated water and in deionized water (control samples), no significant differences were found in sperm mobility, viability, and the number of defective sperm. However, significant differences were noted in the case of samples frozen with plasma-treated water in the presence of aspartate aminotransferase and alkaline phosphatase. The following results were obtained: 152.7 U/L for aspartate aminotransferase and 1612.1 U/L for alkaline phosphatase, while in the control tests, they were, respectively, 187.1 and 2198.3 U/L. According to the authors, fertilization with semen frozen in plasma-treated water was 20% higher, and pregnancy without miscarriages was reported for 73.3% of mothers, compared to 45.0% for samples frozen in deionized water.

Szymanowicz et al. [33] assessed the suitability of plasma-treated water (so-called nanowater) as a solvent for the storage of commercial boar semen. Semen collected from boars was appropriately diluted in plasma-treated water (treated sample) and in non-plasma-treated water (control sample) and stored for 15 days at 16–18 °C. The motility of boar sperm was significantly greater in the samples diluted with plasma-treated water from the 5th to 13th day compared to the control sample (non-plasma-treated water). A decrease in sperm motility (less than 40%) was observed about 24 h earlier in the treated sample compared to the control sample. Plasma-treated water also increased the mean sperm survival time: on the 5th day, the value was 314 min for plasma-treated water, compared to 284 min for the control sample, and on the 10th day, the value was 223 min for plasma-treated water and 182 min for the control sample. These results allowed the researchers to conclude that the plasma-treated water contributed significantly to the prolongation of the lifespan of boar sperm. Other studies showed that watering sows with plasma-treated water increases the effectiveness of their insemination and the maintenance of pregnancy in its first stage, i.e., until the embryos are implanted [34]. A report regarding the use of plasma-treated water for watering chickens [35] showed that using plasma-treated water was related to 14 to 30% fewer falls, compared to the control sample (tap water). Additionally, watering the chickens with plasma-treated water allowed feed consumption to be reduced by approximately 10%. Similar benefits were observed when watering turkeys with plasma-treated water [36]. The number of falls in the group that consumed plasma-treated water decreased by almost 60%, and the average weight of turkeys was higher by approximately 4% in comparison with turkeys watered with non-plasma-treated water. The feed consumption decreased by over 11.5%.

In the food industry, Jung et al. [37] investigated the possibility of using atmospheric pressure plasma-treated water (PTW) as a source of nitrite for curing emulsion-type sausage. The presented results showed that the obtained PTW contained the appropriate amount of nitrite (782 ppm) necessary for the production of the above-mentioned sausages. Used plasma water is not a chemical reagent and is not a natural source of nitrogen. The results showed that plasma-treated water can be suggested as a safe substitute in the meat industry instead of additional chemicals (sodium nitrite) and natural (celery concentrate) auxiliary substances. In another scientific article, Jung et al. [38] focused on investigating the effect of using plasma water as a source of nitrite for meat curing. Plasma-treated water, after plasma treatment for 30 min, contained 46 ppm of nitrite. Meat stuffing was prepared in three different variants (Control, noncured meat batter; PTW, meat batter cured with PTW; Sodium nitrite, meat batter cured with sodium nitrite) to check the effect of PTW on the color of sausages. The presented results confirmed that plasma-treated water can be used as a source of nitrite in the curing process of meat without adding other sources of nitrite. Similar studies were conducted by Young et al. [36] where they used PTW in the production of loin ham.

With regard to plant development, Sitarska [39] proved that the watering of a nonfertilized Tybalt spring wheat crop with plasma-treated water reduced the length of the sprouts of the cultivated wheat by 24% after 7 days and by 1.8% after 14 days. Protein levels slightly decreased, and the levels of chlorophylls a and b remained unchanged. A clearly positive effect was observed during irrigation with plasma-treated water of the fertilized crops; however, in this case, the type of fertilizer used was important. The growth rate when watered with plasma-treated water increased by an average of about 25%. The influences of plasma-treated water on germination, field emergence capacity, the occurrence of other important development stages, as well as the yield and quality of wheat, rye, and string beans, have also been examined [40]. It was found that soaking wheat grain with plasma-treated water significantly influenced the energy and germination capacity. The earlier occurrence of developmental phases was also observed with these seeds. In an experiment with green beans watered with plasma-treated water, an increase in the yield of pods was observed from 54.8 to 69.0%. The first effects were already observed during the plant's vegetative stage. A significant effect of plasma-treated water on seed germination was also observed in the case

of pepper cultivars [41]. Pisulewska et al. [42] investigated the effects of plasma-treated water on the irrigation of peppermint crops. The authors obtained results that showed a significant positive effect of watering with plasma-treated water on the level of essential oils, while their composition also changed. The use of plasma-treated water also increased the content of chlorophylls, carotenoids, and ascorbic acid. Similar results were observed in watercress seeds soaked in plasma-treated water [43]. Research was also conducted on the possibility of using plasma-treated water for the production of plant protection products. This was proven in the studies presented by Jaworska et al. [44,45]. The results showed that fungi and nematodes, known and used as entomopathogenic pesticides, reproduce better after contact with water exposed to low-temperature, low-pressure glow plasma, and their offspring are more pathogenic compared to control samples (without plasma treatment). Zhou et al. [46] focused on the use of water exposed to plasma under atmospheric pressure (PTW) for seed germination and seedling growth of mung bean and its sterilization effect on mung bean sprouts. The results presented in the article showed that the applied PTW contributes to the improvement of seed germination and seedling growth. Scientists also focused on studying the physicochemical properties of PTW in greater detail to better understand the effect of plasma water on improving seed germination. Thanks to their research, scientists have proven that plasma-treated water (PTW) is an environmentally friendly and cheap agent that promotes the germination and growth of mung bean seeds. Very interesting results were obtained during watering *Lavandula angustifolia* with water exposed to a low-temperature, low-pressure glow plasma [47]. In the experiment, water plasma in the air (LPGPA), in oxygen-free nitrogen (LPGPN), in methane (LPGPM), in carbon dioxide (LPGPC), or in molecular oxygen (LPGPO) was used. Plants watered with LPGPN showed a higher protein content and higher magnesium bioaccumulation. The type of water used also influenced the composition of the essential oils. The highest oil content was obtained after LPGPN (0.4 g/100 g dry mass) application, compared to the control sample of nontreated water (0.2 g/100 g dry mass). Plasma-treated water (PTW) was also used as an antimicrobial additive in the washing process of freshly cut lettuce. The obtained promising results could lead to the industrial use of PTW in the processing of freshly cut products to reduce the microbial load on the food surface [48]. Terebun et al. [49] investigated the effect of plasma-treated water on the germination of beetroot (*Beta vulgaris*) and carrot (*Daucus carota*) seeds. The conducted research showed that in the case of beetroot, a positive effect on the number and length of germination seeds was observed, which increased with the extension of treatment time. The influence of RF helium-nitrogen plasma on the germination of onion seeds was also investigated [50]. Presowing plasma stimulation of seeds improved the germination capacity and energy for all the studied groups as compared to the control group. Pawłat et al. [51] in their research investigated the influence of plasma-treated water on the germination process of Thuringian Mallow (*Lavatera thuringiaca* L.). The stimulation of seeds with plasma improved such parameters as: germination capacity and energy for all groups under the study in relation to the control.

The first studies on the possibility of using plasma-treated water in the malting and brewing industries were carried out by Pater et al. [19], who investigated the effect of water treated with low-temperature, low-pressure glow plasma, under an air or nitrogen atmosphere, on the quality of the obtained brewing malt. In this experiment, spring water treated with plasma (treated sample) and untreated spring water (control sample) were used to soak the grain of two-row spring barley. After soaking the grain with plasma-treated water, a much higher water uptake capacity and a greater grain sensitivity to water, as well as energy and germination capacity, were observed. The resulting malt was characterized by a suitable moisture level and thousand-grain weight. The research did not reveal any differentiating effect of the tested malts throughout the course of the beer wort production processes.

Plasma-treated water is also used in cosmetology, due to its superior ability to dissolve mineral salts [52], and in medicine. Ohl et al. [53] proved that water treatment by plasma can be used to improve various therapeutic properties of medical implants. Another

possibility of using low-temperature, low-pressure glow plasma is in the preparation of vascular prostheses, the purpose of which is to release a drug that reduces blood clotting and thrombosis [54]. Schloser et al. [54] described the synthesis of biomaterials. Currently, there are studies checking the use of low-pressure, low-temperature plasma in cleaning the environment [55,56]. Much attention has been paid to the possibility of purifying water from impurities by means of plasma [57]. In turn, studies carried out by Schnabel et al. [58] investigated the synergistic antimicrobial activity of plasma-treated air and water in the decontamination of surfaces and food processing environments. In this paper, researchers focused on proposing a new cleaning and disinfection procedure based on a single source of antimicrobial agent. In the article entitled “Treatment of surface water using cold plasma for domestic water supply” [59], the authors presented the results of using cold plasma for surface water treatment. The obtained water was used for domestic use. This is another article that confirms that cold plasma can be effective in destroying bacteria in water. The authors also noticed that the plasma process contributes to an increase in the content of nitrites and nitrates in the water.

There is increasingly more interest in the use of plasma water to inactivate microorganisms. Shaw et al. [30] in their research examined the inactivation of bacteria by plasma-treated water (PTW) enhanced by reactive nitrogen species. In these studies, the authors focused primarily on investigating the role of the reactive form of nitrogen (RNS) as an antibacterial agent. The obtained results showed that  $N_2 + 0.5\% HNO_3$  vapor plasma was most effective in inactivating bacteria, compared to the PTW generated by plasma without the vapor system. Kitano et al. [60] focused on the extraction of bactericidal components in cryopreserved plasma-treated water. In 2017, research was carried out on the possibility of using plasma-treated water (PTW) as an insecticide. Bosch et al. [31] investigated the effect of such water on mealybug (*Planococcus citri*). Plasma-treated water was characterized by a lower pH. After 10 min of treatment, the researchers obtained water samples with a pH value of 1.8. It was noticed that, compared to the classically acidified waters with nitric and hydrochloric acid, an approx. 90% higher mortality of mealybugs was shown after using PTW. Similar studies were carried out by scientists from Germany, who focused on investigating the effect of plasma-treated water (PTW) on the removal of biofilm induced by *P. fluorescens* [61]. In their next article, scientists focused on combining plasma-processed air (PPA) and plasma-treated water (PTW), resulting in the synergistic inactivation of *Candida albicans* SC5314 [62]. Pawlat et al. [63] investigated whether municipal waste used for the production of alternative fuels can be effectively sterilized with low-temperature, low-pressure glow plasma (LPGP) to prevent water contamination and threats to workers' health.

## 5. Conclusions

The use of low-temperature, low-pressure glow plasma (LPGP) for the production of plasma-treated water is a promising method that may be commercialized in the future. This technology is supported primarily by the results of research carried out in plant breeding, animal husbandry, cosmetology, medicine, and the food industry. Initially, water was treated with low-temperature, low-pressure plasma in the presence of air, which stimulated various microorganisms, aided plant growth, and ensured the healthy breeding of selected animals. On the other hand, water obtained by treatment with plasma (LPGP) under anaerobic nitrogen may be used for the cultivation of anaerobic microorganisms and the growth of plants using atmospheric nitrogen. Such studies have prompted scientists to produce plasma-treated water in the presence of ammonia. The obtained water protected heat exchangers and pipelines against the formation of limescale. Research has also been carried out on water plasma in the presence of methane. The analyses of Raman spectra suggested the formation of specific aqueous methane clathrates.

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