

Interaction of atmospheric pressure plasma with a liquid electrode

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This research focuses on the interaction between liquid electrode (water) and atmospheric discharge streamers in air. It elucidates water surface deformations due to electrohydrodynamic effects in high-voltage discharge-distilled water interaction. The aim is to comprehensively study processes in the water layer under electrical discharges, with research areas covering discharge parameter measurement, water surface deformation control, and electrode change analysis. The study holds scientific significance in enhancing understanding of electrohydrodynamic effects and plasma-liquid interaction mechanisms. Practically, it has potential applications in water treatment, liquid purification, and materials science. Research methods involve analyzing electrical characteristics of the discharge and surface properties of a metal grounded electrode in water. Key results show water surface deformation, electrolysis, and discharge electrical property changes at a fixed liquid thickness, characterizing plasma-liquid interaction dynamics and enabling optimization of related technologies. The results can enhance water treatment and purification, assist in new material and coating development. In water treatment and liquid purification, they can boost pollutant decomposition via plasma and electrolysis. In materials science, they can guide new anticorrosive coating development on metals by studying oxidation and deposition under plasma discharge. The data also benefits other plasma-technology-related fields.

Key words: plasma-liquid interactions, discharge in water, streamer dynamics, deformed water surfaces.

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1. Introduction

The non thermal gas discharges plasmas are the source of various charged species, metastable atoms, active radicals, UV photons and widely applied in the field of nanomaterial synthesis, surface treatment, gas conversion, agriculture, medicine and wastewater treatment [1-5]. The atmospheric pressure non thermal plasma sources are attracting more attention due to the high versatility, scalability and exhibit various interesting physical phenomena such as microdischarge [6,7] and self-organized pattern formation. In the field of plasma-liquid interaction, many studies have addressed various aspects of this physical phenomenon. Cavendish's work [8] was one of the first in this area, focusing on the production of nitric acid using electric sparks in air. These experiments were crucial for understanding the chemical reactions that occur in plasma-liquid interactions

(“Experiments on Air”). Subsequently, scientists shifted their attention to a deeper understanding of chemical processes [9-11], the study of electrolysis [12-14], electrical destruction of dielectric liquids for high-voltage switching, and the interaction of plasma with liquids for applications in environmentally friendly technologies [15].

Some more recent works focus on effects such as various deformations of the water surface and electrohydrodynamic effects, which are widely used in various applications and electrospray technologies.

The work of Kawamoto and Umezu [16] provided a deeper understanding of the effects of corona discharge on the pressure and deformation of the water surface, emphasizing the importance of linking these effects to the so-called “ionic wind” generated by the discharge. Research in the field of electrohydrodynamic instability conducted in [17] proposed a new method for analyzing the data of

instabilities generated by corona discharge in dielectric liquids. This research explores their effect on surface and volume deformations and considers the properties of the discharge and its potential for application in technical devices.

The changes in the water surface in metal-water discharges have been carefully studied in [18], allowing the determination of different deformation modes depending on the discharge parameters. These studies provided key insights into the behavior of discharges and their potential in various applications.

Other research, such as [19], has provided important data on the effects of corona discharge on the surface characteristics of thin films of various liquids, as well as on the study of the self-sustaining plasma Moses effect. The results of [20] represent a detailed study of a discharge in the atmosphere and above the surface of water under conditions of exposure to nanosecond pulses of high voltage. The study of the decomposition of polystyrene upon contact with water in [21] also provided valuable insights into the applicability of the discharge in chemical synthesis processes.

Although the field of plasma-liquid interaction research is promising and broad, it includes many unresolved aspects that require further study and attention. In particular, it is important to understand the processes that occur during the interaction of a discharge with a liquid and the changes that occur in both the plasma and the liquid during this interaction. Some processes at the plasma-liquid interface remain unknown. Therefore, these studies are also important in fields such as medicine, environmental science, materials synthesis [2,22], and analytical chemistry [12,23].

In this context, our research aims to study the physical processes involved in the exposure of atmospheric pressure plasma to liquid electrodes (with low conductivity) and their influence on the physical properties of the discharge. Particular attention is paid to the occurrence of electrical breakdown at the air-liquid interface, the physical phenomena that occur during these interactions, and their features and mechanisms of formation. These aspects are key to understanding the mechanisms of interaction between atmospheric pressure discharges and liquid electrodes.

In previous studies on the interaction of plasma with water, in many cases the emphasis was placed

on explaining chemical processes, and some works like ours used plasma flow in their research.

Unlike the above-mentioned works, our research focuses on the interaction of atmospheric pressure plasma with low-conductivity liquid electrodes and explains the deformations that occur on the water surface during this interaction without the use of plasma flow. In addition, electrical breakdown and electrolysis processes in such a system are being investigated, which expands the understanding of the mechanism of interaction in the plasma-liquid system.

Thus, our work contributes to the deepening of knowledge about the physical processes occurring at the plasma-water interface and their effect on discharge characteristics.

The purpose of our research is to study the physical processes involved when atmospheric pressure plasma interacts with a low-conductivity liquid electrode (a layer of distilled water) and the mechanisms of plasma action on the surface of a metal grounded electrode in a liquid medium.

To achieve this goal, we used experimental methods to analyze the electrical characteristics of the discharge at a fixed thickness of a low-conductivity liquid electrode. We also examined changes on the surface of a copper electrode in a liquid medium to understand the processes resulting from the propagation of a discharge through a “discharge-liquid-metal” medium.

2. Methods

The experimental setup shown in Figure 1. It consists of an electrode system including an upper and lower electrode, a power supply, oscilloscope, voltage divider, and resistor.

At the bottom of a quartz vessel with a closed window, there is a grounded lower electrode covered with aluminum. On top of this, a copper plate with a thickness of 2.9 mm and a diameter of 25 mm is placed, serving as the cathode.

The upper electrode is located 9 mm above the surface of the copper plate ($l_{u.e.} = 120 \text{ mm}$, $d_{u.e.} = 3.75 \text{ mm}$, $r_{tip} = 0.27 \text{ mm}$), made of stainless steel (AISI 304) with a pointed tip, which serves as an anode for high voltage discharge.

The upper electrode is connected to a sinusoidal high-voltage power supply (PVM-500), after which

the voltage signal is fed through a divider Tektronix P6015A ($1000\times 3.0\text{pF } 100\text{M}\Omega$) to the SIGLENT SDS 1204X-E (200MHz, Dual 1GSa/s, Quad 500MSa/s) for measuring current and

voltage waveforms. One of the oscilloscope probes is connected to a low-inductance resistor ($R=57\Omega$, 10W) for further data processing and analysis of data based on the measured current and voltage values.

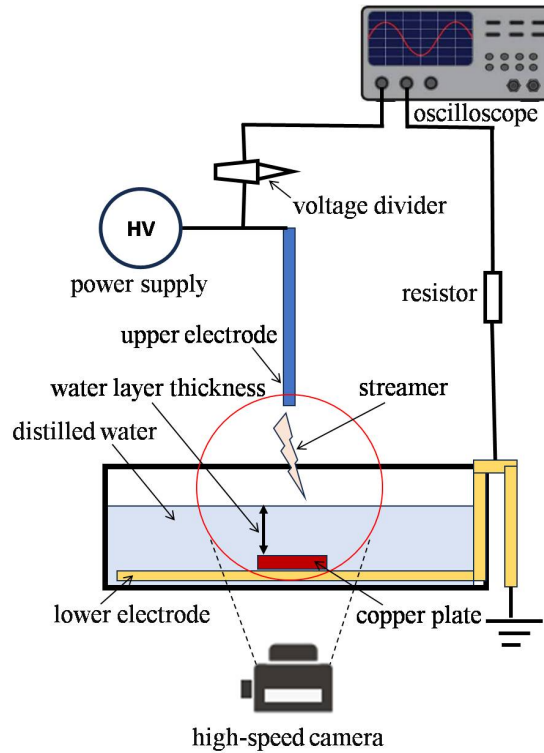


Figure 1 – Schematic of the experimental setup

A 5 mm thick layer of distilled water is poured into a quartz vessel measuring $4.5 \times 4.9 \times 4.5$ cm. Distilled water, with its low conductivity of $18.2 \mu\text{S/cm}$, acts as a good insulator.

A photographic image of the discharge was captured using the PhantomVEO710S high-speed camera, operating at a frequency of 20,000 frames per second. The exposure time for each frame is $49 \mu\text{s}$.

Using an TOPDON ITC629 infrared thermographic camera (Frame Rate of Thermal Images 9Hz), the temperature of the liquid and the top electrode is measured.

The main parameters of the experiment: $p = p_{atm}$, $f = 27 \text{ kHz}$, $t_{d.i} = 5 \text{ min}$ (discharge ignition time) (Table 1). Water thickness: $h_{water} = 5 \text{ mm}$, interelectrode distance: $h_{air} = 4 \text{ mm}$, $h_{copper \text{ plate}} = 2.9 \text{ mm}$ (Figure 2).

Raman spectroscopy was used to analyze the surface of the copper electrode after the experiments. The measurements were carried out using a laser with a wavelength of 473 nm, which provides high sensitivity to fluctuations in copper-oxygen bonds. The spectral resolution of the device was 4 cm^{-1} , which makes it possible to accurately identify peaks and their correspondence to certain vibrational modes.

Table 1 – Parameters before and after the experiment

Parameters	$h_{water}, \text{ mm}$	$t_{water}, ^\circ\text{C}$	$t_e, ^\circ\text{C}$
Before the experiment	5	22.3	22
After the experiment	4.6	39	48

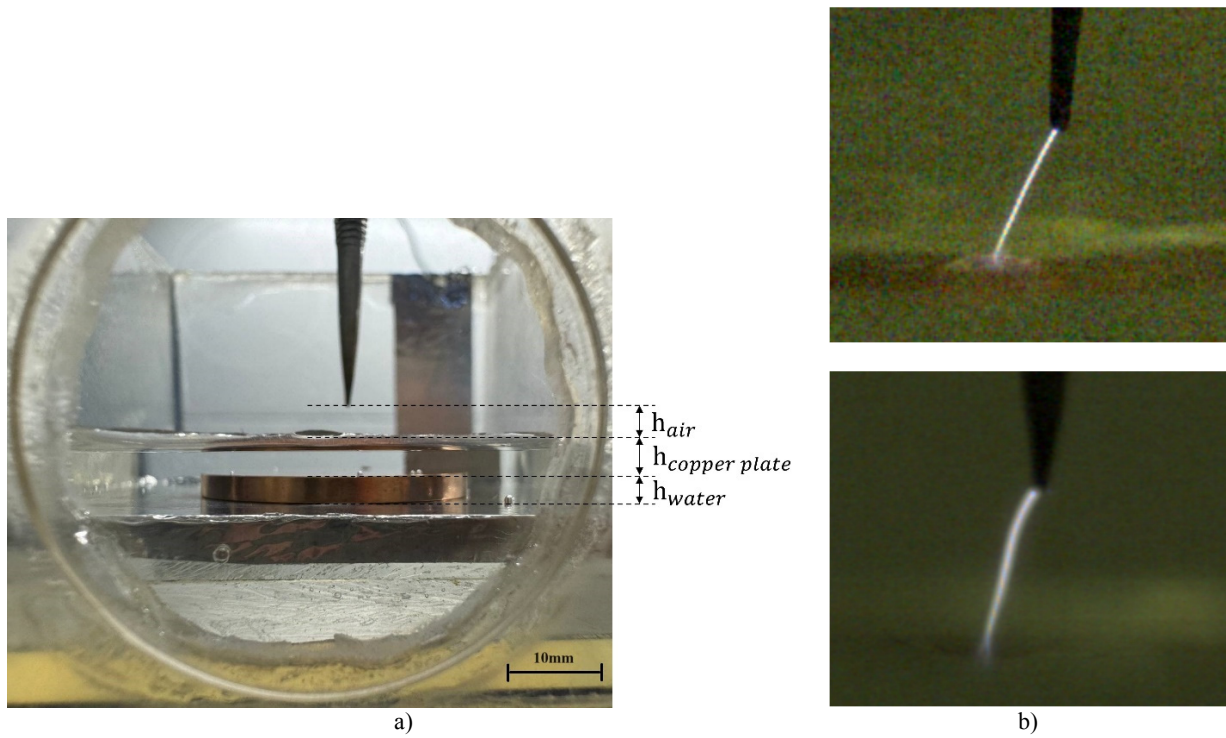


Figure 2 – a) electrode location, b) type of plasma

3. Results and discussion

During the experiment, voltage was applied to the upper electrode. This resulted in the formation of an electric field between the tip of the upper electrode and the surface of the water, within the layer of air. We will refer to this distance as the interelectrode distance.

We can visually observe the occurrence and propagation of a discharge within a layer of air, and the deformation of the water surface was captured using a high-speed camera.

As a result of the experiments, several processes were discovered: deformation of the water surface, the formation of sediment on the copper electrode and electrical breakdown. Let's consider these phenomena separately.

One of the observed phenomena during the experiment is the deformation of the water surface during the interaction with discharge streamers. Figure 3 displays photographs of experiments with water at a fixed thickness. As depicted in the figure, the deformation of the water surface is evident: the discharge induces depressions on the surface of the water.

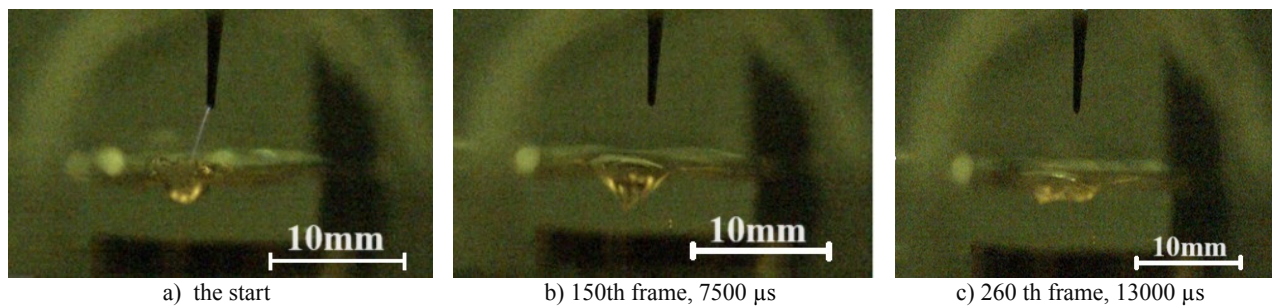


Figure 3 – Deformation of the water surface obtained with the PhantomVEO710S high-speed camera
sample rate 20000 f/s, exposure time 49 μ s, $t_{d.i} = 5 \text{ min}$ (discharge ignition time),
 $h_{\text{water}} = 5 \text{ mm}$, $h_{\text{air}} = 4 \text{ mm}$, $h_{\text{copper plate}} = 2.9 \text{ mm}$

The deepening of the water surface occurs due to the electrohydrodynamic force [9,10]. In our discharge, ions are accelerated in one direction under the influence of an electric field. As they collide with neutral atoms in the air, and neutral atoms subsequently collide with the surface of the water, deformation occurs. This phenomenon is known as the electrohydrodynamic effect [24].

Deformation of the water surface commences upon the application of voltage. As the discharge voltage gradually increases, the intensity of the electromagnetic field rises, reaching a peak, resulting in a maximum deformation depth of about 2-3 mm (Figure 3a). However, as the discharge or voltage decreases, the electromagnetic effect diminishes, and the water molecules on the surface return to their original positions due to surface tension. This gradual process leads to the surface calming down and the disappearance of deformation (Figure 3b, c).

Figure 4a displays an oscillogram depicting the variation of voltage and current in time at a fixed thickness of the distilled water layer. This graph covers the whole process, starting from the very beginning of the experiment, and allows us to observe the general character of the changes, as well as to identify the moment of electrical breakdown.

Figure 4b shows the part of the graph focused on the moment of electrical breakdown. This plot shows sections of the half-periods at which the electrical breakdown occurs, and three half-periods near the breakdown are obtained. This allows us to examine the breakdown process and the changes in voltage and current in the vicinity of the breakdown point. According to the obtained data, electrical breakdown occurs at $t = 201\mu\text{s}$, with a breakdown voltage of $U_{br} = 6,5\text{ kV}$ ($I = 87,7\text{ mA}$).

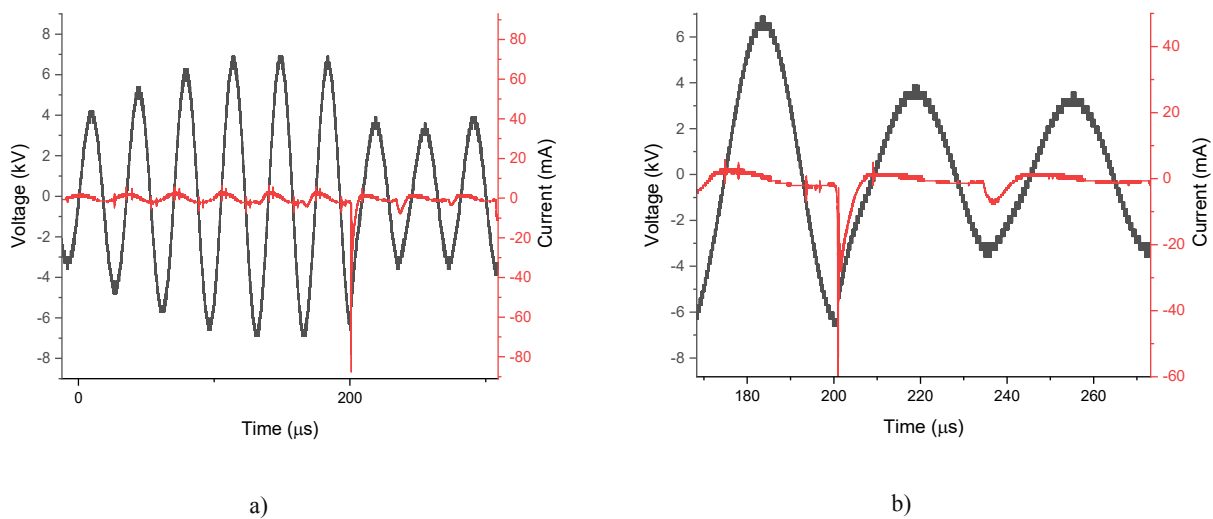


Figure 4 – Instantaneous current and voltage waveforms
 $p = p_{atm}$, $f = 27\text{ kHz}$, $t = 22^{\circ}\text{C}$

Another process discovered after the experiment is the deposition of thin film on the surface of the copper plate. A solid dark spot formed in the center of the copper disk, and small spots were observed along the edges. Analysis by optical spectroscopy revealed that between the black spots in the center of the copper plate, there were slightly concave yellow spots. Additionally, black spots were observed all over the surface, except for the center of the plate, among which yellow spots were also present in the

middle (Figure 5). These stains are the result of electrolytic processes, so we will further discuss their nature and analysis.

When a high voltage is applied, air ionization occurs between the tip of the upper electrode and the surface of the water. This process leads to the formation of a plasma channel, which serves as a conductor for charged particles (ions and electrons), allowing them to pass between the electrode and water. Once the plasma channel is installed, the

electrons moving through it reach the surface of the water and initiate the electrolysis process. At the moment of discharge, a local electric field is formed

on the surface of the water [25], which leads to charge separation between the cathode and the anode, thereby contributing to electrolytic reactions.

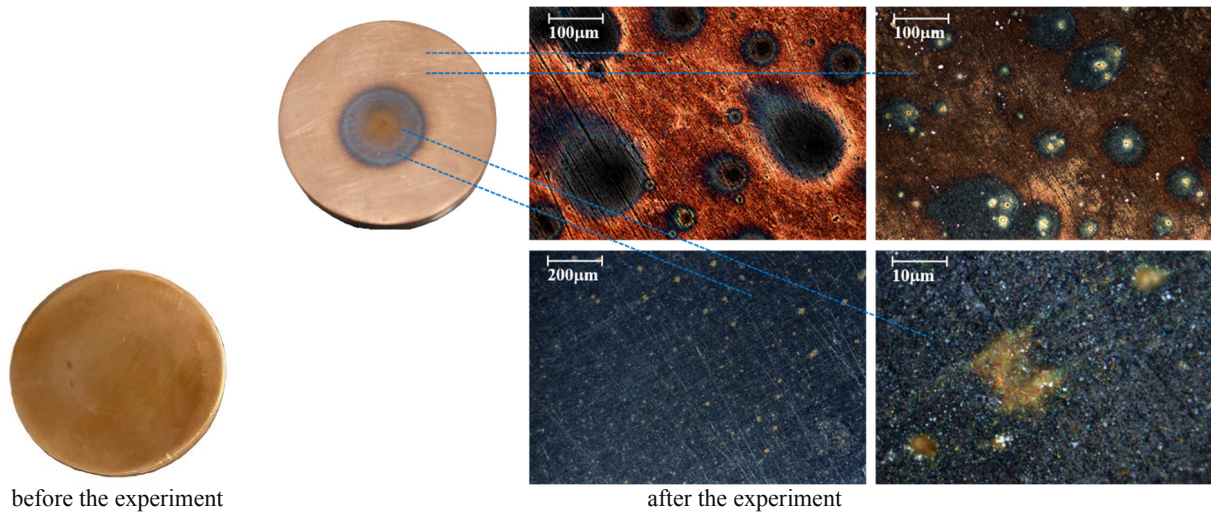
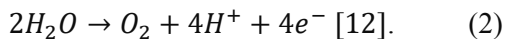
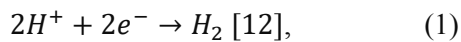


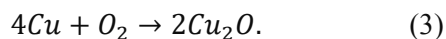
Figure 5 – Edge and centre of the copper plate (after the experiment)

$t_{d.i} = 5 \text{ min}$ (discharge ignition time), $h_{\text{copper plate}} = 2.9 \text{ mm}$, $d_{\text{copper plate}} = 25 \text{ mm}$, $d_{\text{spot}} \approx 10 \text{ mm}$

Under normal conditions, plasma-liquid interaction induces a sequence of chemical reactions [4], including the electrolysis of water, leading to the formation of hydrogen and oxygen at the electrodes. Therefore, this system can be characterized as water electrolysis with sufficiently high potential, involving electrolytic reactions at the cathode and anode. Electrolysis of water involves two primary reactions: reduction process at the cathode (-) (1) and oxidation process at the anode (+) (2):



During the electrolysis of water, the copper surface reacts with oxygen to form various compounds such as copper oxides or hydroxides can be formed. This is due to reactions of copper with products of water electrolysis such as oxygen or hydrogen. The reaction at the anode produces oxygen, which can interact with the copper surface, leading to the formation of copper (II) oxide:



Thus, the formation of copper (II) oxide can lead to changes in the color and structure of the surface, which appears as stains.

To fully understand the processes occurring and determine the nature of the resulting compounds or structures, the surface of the copper plate was analyzed using Raman spectroscopy. In the spectra taken from the center and edges of the copper plate, characteristic peaks at 147.7 , 213.7 and 642.7 cm^{-1} were revealed, which correspond to copper oxide(I) – cuprite (Cu_2O). These peaks indicate the presence of fluctuations in copper-oxygen bonds in the cuprite structure:

- 147.7 cm^{-1} is the peak corresponding to the oscillation of the copper–oxygen bond in the cuprite structure (oscillatory mode A_{1g});

- 213.7 cm^{-1} is a peak that indicates symmetrical oscillations in octahedral clusters characteristic of $\text{Cu}_2\text{O}(\text{Eg})$;

- 642.7 cm^{-1} – indicates deformation fluctuations of the $\text{Cu-O}(\text{T}_{1g})$ bond.

The spectrum (Figure 6) indicates the presence of copper (I) oxide or cuprite (Cu_2O) with an oxidation state of +1 (I), both at the edges and in the center of the plate. This indicates that

each copper atom in this compound carries a positive charge of +1. The presence of cuprite on the surface of the copper plate was confirmed by

comparing the peaks of the Raman spectra with those from other studies, including those provided in the cited article [26].

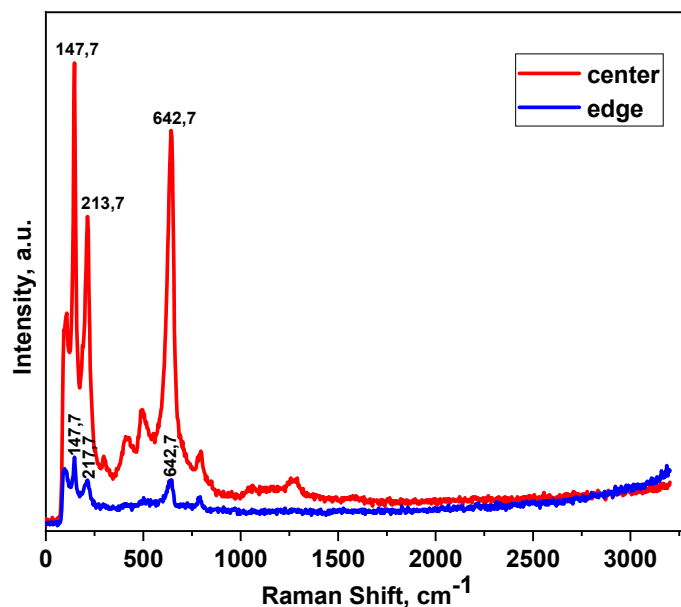


Figure 6 – Results of Raman spectroscopy

Thus, the results of Raman spectroscopy and visual observations show that cuprite (Cu₂O) and, probably, copper (II) oxide (CuO) are formed on the surface of the copper electrode. Their distribution and quantity depend on the intensity and duration of the discharge. With prolonged discharge, the accumulation of electrolysis products and the interaction of copper with plasma on the surface of the plate increases, which contributes to the formation of more cuprite and other oxidized compounds. In addition, at higher voltage and discharge intensity, more active particles are generated, which enhances the oxidation of copper. In the center of the plate, where the discharge conditions are most intense, deeper oxidation processes are observed, which is confirmed by the appearance of dark spots. These results demonstrate that changes in the composition and structure of deposition on the electrode are directly related to the duration and parameters of the discharge.

4. Conclusion

In this paper, we offer an explanation regarding the observed phenomena that occur during the

interaction of atmospheric pressure plasma and distilled water. It has been determined that the deformation of the water surface during the operation of a discharge is related to the action of the electrohydrodynamic force. As the voltage rises, the depth of the water surface deformation also increases, and this deformation vanishes when the discharge is switched off. A graph illustrating the electrical breakdown during the interaction of water and discharge was presented. At a discharge time of $t = 201\mu\text{s}$, an electrical breakdown takes place; through the use of an oscillogram, it was established that the breakdown voltage is 6.5 kV.

During the experiment, a distinct spot appeared on the surface of a copper plate following the application of a discharge. This phenomenon is attributed to electrolysis. The series of reactions resulting from the interaction between water and discharge is described: it involves cathodic reduction and anodic oxidation reactions, which ultimately lead to the formation of copper oxide when the copper surface interacts with the reaction products. Raman spectral analysis of the copper plate surface confirmed the presence of copper oxide, as indicated by prominent peaks. This observation indirectly

validates that the characteristic spots on the copper plate surface are a consequence of electrolysis rather than the direct propagation of discharge through an aqueous medium.

The observed effects define the dynamics of the interaction between plasma and liquid. The results obtained enable us to gain a more profound understanding of the interaction between plasma and liquid and to develop novel technologies based on these interactions.

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