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Low-cost Cold Atmospheric Plasma Source for Surface Sterilization

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Resumo. *Plasmas possuem diversas aplicações na indústria, muitas delas no tratamento de superfícies. Este trabalho explora as propriedades de Plasmas Frios Atmosféricos para a esterilização de superfícies, objetos e ambientes. Uma fonte de plasma atmosférico foi desenvolvida utilizando diferentes topologias de circuitos, com sinais AC ou pulsos DC. Então, um protótipo utilizando a topologia de pulsos DC foi construído, com materiais de baixo custo. Resultados preliminares da ação biocida do plasma, utilizando o* Peribacillus simplex*, demostraram que a fonte de plasma é capaz de esterilizar após 10 minutos de exposição.*

Palavras chave: *plasma frio atmosférico, fonte de alta tensão, esterilização por plasma, circuito pulsado DC, tranformador flyback*

Abstract. *Plasmas have several applications in industry, mainly in surface treatment. This work aims to apply the properties of cold atmospheric plasmas for the sterilization of surfaces, objects, and environments. An atmospheric plasma source has been developed using different circuit topologies that work with high frequencies, on the order of KHz, utilizing AC or DC-Pulsed signals. Then, a prototype DC circuit was implemented employing inexpensive materials. Preliminar results of the biocide action using* Peribacillus siplex *demonstrate that the plasma source is able to sterilize after 10 minutes of exposition to the plasma.*

Keywords: *cold atmospheric plasma, high voltage power supply, plasma sterilization, DC-pulsed circuit, flyback transformer*

1. INTRODUCTION

Surface treatment using plasmas is a process widely used in industry. Among the applications we can mention the deposition of thin films, the removal of material through the sputtering (etching), the welding of metals, the doping of semiconductors, electric thrusters, fabrication of nanomaterials, and, among other diverse uses, plasmas can be used to eliminate biological contaminants.

There are different ways to sterilize medical equipment, for example, using ovens, antiseptics, disinfectants, autoclaves, and ultraviolet, among others (Campos de Souza, 2012; Kaushik *et al.*, 2023). Each process has its characteristics that make it suitable for certain applications, and inappropriate in other situations.

The application of plasmas for the elimination of contaminants can be classified into vacuum processes and atmospheric processes. Vacuum processes use a closed chamber and a low-pressure environment to ionize a specific gas, such as hydrogen peroxide. The resulting plasma fills the entire internal space of the chamber, through diffusion, and the constituent ions of the plasma react with biological contaminants, performing sterilization. Atmospheric processes can be applied in open and closed environments, at atmospheric pressure, ionizing the atmospheric air, and producing a cold plasma. The reactive ions are capable of killing microorganisms such as bacteria and fungi and deactivating viruses. The effectiveness of atmospheric processes depends on the duration of exposure to the ionized gas (Izadjoo *et al.*, 2018). This plasma generated from atmospheric air, and at pressures close to 1 atm, is known as Cold Atmospheric Plasma (CAP).

With the recent SARS-CoV-2 pandemic, the need arose to obtain different and affordable ways to sterilize surfaces, objects, and environments in a safe and reliable way. In recent years, several applications for atmospheric plasmas have been studied and developed, with a focus on eliminating the virus that causes COVID-19. This concept has been applied to the treatment of objects and surfaces (Moritz *et al.*, 2020), such as PFFF3 masks (Schmidt *et al.*, 2021), as well as in the sterilization of environments, wounds, dressings, and medical equipment (Izadjoo *et al.*, 2018; Chen *et al.*, 2022). This scientific effort also brought other applications for CAPs, such as in food preservation processes (Yinxin *et al.*, 2022) and in the manufacturing process of medical materials (Moritz *et al.*, 2020).

Plasmas can inactivate biological contaminants through different mechanisms, such as the production of free radicals, reactive nitrogen species (RNS), reactive oxygen species (ROS), electric fields, charged particles, and ultraviolet (UV) photons (Chen *et al.*, 2022; Kaushik *et al.*, 2023). These mechanisms can act directly, or indirectly, on sterilization, for example, the production of free radicals and reactive species can act directly on biological contaminants (through the diffusion of gas around the object to be sterilized), or by dissolving these chemical compounds in a liquid solution to be used as an antiseptic cleaning material (Chen *et al.*, 2022; Kaushik *et al.*, 2023).

The reactive species in CAPs can be: atomic oxygen, hydroxyl radicals, hydrogen peroxide, superoxide, and ozone. They interact with the liquid medium and can create different species in the plasma, plasma-liquid interface, and liquid medium.

According to Kaushik *et al.* (2023), among the chemical reactions in CAPs for the production of reactive species, we can highlight:

- Atomic Oxygen:
  $M^* + O_2 \rightarrow M + 2O, (M : He, Ne, Ar)$
  $M_2^* + O_2 \rightarrow 2M + 2O$
  $e^- + O_2 \rightarrow O + O + e^-$

- Hydroxyl Radical:
  $M^* + H_2O \rightarrow M + H^\bullet + O^\bullet H, (M : He, Ne, Ar)$
  $M_2^* + H_2O \rightarrow 2M + H^\bullet + O^\bullet H$
  $e^- + H_2O \rightarrow O^\bullet H + H + e^-$
  $UV + H_2O \rightarrow H_2O^*$
  $UV + H_2O^* \rightarrow H^\bullet + O^\bullet H$
  $H_2O^* \rightarrow OH^- + H^+$
  $OH^- \rightarrow O^\bullet + e^-$

- Hydrogen Peroxide:
  $e^- + O_2 \rightarrow O_2^-$
  $O^\bullet H + O_3 \rightarrow HO_2 + OH^-$
  $O_2^- + H_2O \leftrightarrow HO_2 + OH^-$
  $OH + OH \rightarrow H_2O_2$
  $H_2O_2 + OH \rightarrow HO_2 + H_2O$

- Ozone:
  $O + O_2 + M' \rightarrow M' + O_3$

There are many mechanisms by which these different elements, such as UV radiation, heat, gamma radiation, reactive oxygen species (ROS), reactive nitrogen species (RNS), and reactive oxygen-nitrogen species (RONS), affect biological contaminants. They can effectively kill biological contaminants by membrane lipid peroxidation, plasmid DNA damage, chromosomal DNA/RNA damage, induced oxidative stress, protein denaturation, and amino acid oxidation.

The Plasma Physics Laboratory at the University of Brasilia (LFP/UnB) has been developing technological applications based on plasmas since 2002, including a plasma reactor for sterilization at atmospheric pressure (Campos de Souza, 2012). A dielectric barrier discharge was obtained from atmospheric air and an alternating current circuit with 16.3 kV and 60 Hz. The biocidal action was characterized using spores of *geobacillus stearothermophilus*, and no remaining viable spores were detected after 40 minutes of plasma exposition.

In this paper, we present the details of a CAP source prototype constructed using inexpensive and easily available materials. Low-cost solutions were a central pillar in the design process, leading to some choices along development, for example, the use of DC flyback transformers from old CRTs, which in turn guided the DC circuit topology. The same can be said about disposable syringes, proving their endurance as an insulator, structural piece, and plasma confinement at reduced cost. Also, by utilizing a modified discharge model of Neon lamps, we could simulate different circuits and component values, obtaining an approximate behavior as the real discharge. Further refinement of this simulation model and comparison with real discharge data, utilizing a 20KV oscilloscope probe, will be presented in a future work. Preliminary results of the biocidal action are also presented, demonstrating the ability of the prototype for surface sterilization. For this, samples of *Peribacillus Simplex* were submitted under a CAP discharge, and after incubation, the results were compared with duplicate controls.

This paper is organized as follows. Section 2 describes different techniques to obtain CAPs. Section 3 presents the circuit topology employed to implement the CAP source, with SPICE simulations and a prototype circuit. Section 4 brings the preliminary results, and Section 5 the conclusion and final considerations.

2. COLD ATMOSPHERIC PLASMA SOURCES

Cold atmospheric plasmas sources can be obtained with different electrical discharges such as corona discharge, dielectric barrier discharge, and glow discharge. These types of plasma can contain electrons, UV photons, ions, neutral atoms, and molecules (Chen *et al.*, 2022). A cold plasma jet can be obtained from a glow discharge. This jet is an extension of the plasma density decay from the generation point (Chen *et al.*, 2022; Zhu *et al.*, 2021). Thus, by regulating the electrical parameters of the discharge and the flow of the gas, we can control the parameters of the jet, such as density, intensity, and plume size. These devices are usually pen-shaped, with a dielectric material serving as the structural body, an electrode on the inside of the tube, and an electrode on the outside of the tube. The gas flows through the electric field generated by the electrodes and is ionized, resulting in a plasma plume at the exit of the tube.

Corona-type plasma discharges use electric fields capable of ionizing the gas around an electrode, but without allowing arcing or dielectric breakdown to occur (Zhaoquan *et al.*, 2020; Chen and Wirz, 2020). These electric fields can be continuous (DC) or alternating (AC) and the current drawn by the discharge is around micro and a few milliamps. Glow-type plasma discharge has a higher current density than corona discharge, but lower than arc discharge. Usually glow discharges vary from a few hundred microamps to one amp.

Atmospheric corona and glow discharges require high voltage and low current. The electric arc in these systems uses a low voltage and high current. The electric arc also has applications in the generation of CAPs and in different industrial processes, such as the generation of ultra-fine particles, cutting, welding, waste treatment, and plasma spraying, among others.

3. HIGH VOLTAGE AC AND DC PULSED CIRCUIT

Different topologies of electronic circuits can be used to carry out the ionization of atmospheric air, and thus promote the elimination of biological contaminants from surfaces, environments, and objects. In this section, we describe two topologies, namely, Zero Volt Switching and DC Pulsed.

Zero Volt Switching (ZVS) consists of an oscillating circuit where the activation of one transistor causes the deactivation of the other, thus generating an oscillation of voltage and current in the inductor, which may be the primary of a transformer. The frequency in which this circuit oscillates is determined by the relationship between the inductance (primary of the transformer) and the capacitance in parallel. This circuit can supply current pulses of the order of tens, and even hundreds, of amperes to the primary of the transformer, making it possible to work with high powers of the order of hundreds, even thousands, of watts. One application of these high-current pulses is seen in induction heaters. An important aspect of this circuit for the ionization of atmospheric air, in addition to the possibility of working with higher powers, is the AC nature of the output signal, alternating polarity each time the oscillator passes through zero volts.

DC Pulsed is a circuit that uses pulses from a DC signal on the primary of the transformer to create high voltage pulses on the secondary. These pulses can be in the form of a pulse-width modulation (PWM) signal (square wave) that allows the control of the frequency and pulse width of the generated signal. A possible example of this topology consists of a PWM signal generator circuit controlling a power stage, made up of transistors (Thana *et al.*, 2019) and (Pei *et al.*, 2018). These, in turn, are responsible for switching the primary of the transformer, creating voltage and current pulses in it. In addition to signal adjustment, these circuits allow the control of the power delivered to the transformer (and in turn to the plasma), allowing operation with varying levels of power and avoiding the transition to an electric arc.

In this work, a test circuit was developed using the DC Pulsed topology. A PWM signal generator, using a 555 IC and an LM741 operational amplifier, was coupled to a MOSFET (IRFZ44N), in order to create pulses in the primary of a FLYBACK transformer, model OV20762F, taken from a CRT television. With this circuit we can control the DUTY-CYCLE of the PWM signal, and its frequency, thus adjusting the consumed power of the source and the intensity of the corona discharge. The plasma wand was built with a 5 mL hypodermic syringe, which was modified to allow air to enter through the upper part, and the positive electrode to pass through. This circuit allows the adjustment of the discharge to find the optimum operating point for the system while using inexpensive materials. The total cost of the system is estimated at approximately 20 dollars at the time of writing.

Figure 6 displays the prototype mounted on a universal perforated board. The power supply for this circuit was carried out with a bench source (model PS-1502DD), supplying 15 V and 1.2 A approximately, which is equivalent to 18W. The circuit can also be operated with a 12V battery.

The corona discharge between the plasma wand and the ground plane connected to the ground of the high-voltage transformer is shown in Fig. 7. Figure 8 shows the discharge being applied to the substrate of a Petri dish, for the tests to eliminate biological contaminants.

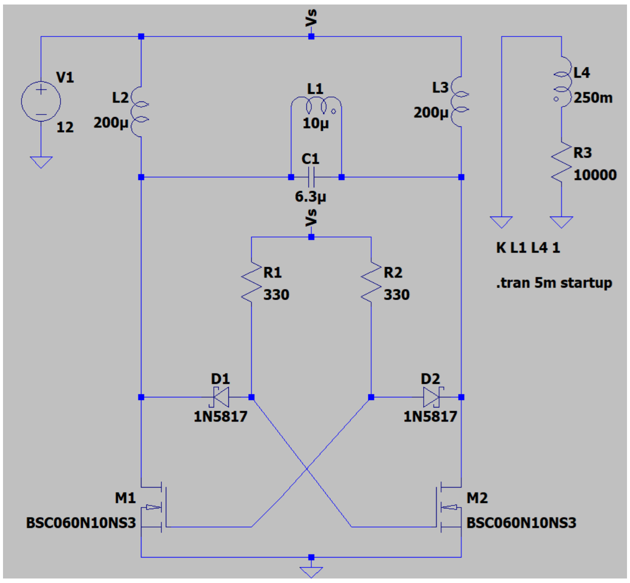

Figura 1. Simplified circuit of the bistable oscillator used for the simulation in LTSPICE. This topology is also known as Zero Volt Switching (ZVS).

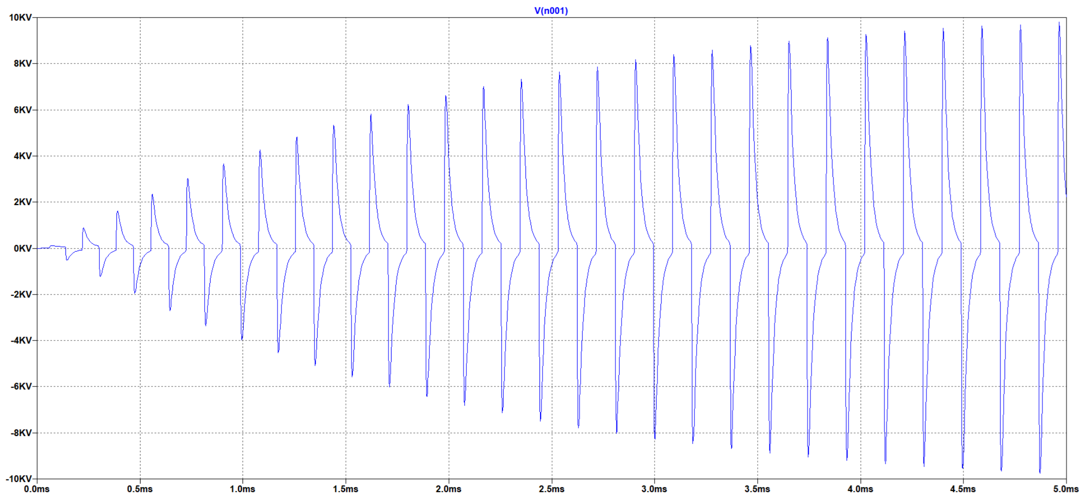

Figura 2. LTSPICE simulation result:It is possible to observe the building up of tension in the output of a flyback transformer. This simulation didn't account for the discharge dynamics of plasma, only the possibility of achieving voltage potentials necessary for gas ionization. This circuit achieve a steady state around 10kV.

3.1 LTSPICE Simulations

The free software LTSPICE XVII was chosen to simulate and comprehend the different topologies and limits of operation, such as current draw, NMOS power dissipation, frequencies, and what components to use in the prototype circuit.

We start by simulating the ZVS topology, where the frequency is controlled by the relation between capacitors and inductors (such as the transformer's primary coil). Figure 1 illustrates the ZVS circuit schematic. Figure 2 and Fig. 3 show the output voltage and resonating frequency.

A positive aspect of this circuit is the self-regulating resonant frequency, which has the capability to make minor adjustments due to the naturally varying impedance of a plasma. A downside is the lack of controllability of the discharge parameters and the higher input current characteristic of this circuit. Although this higher input current allows for higher currents in the discharge, it elevates the cost due to the necessity of sturdier components. Also, the flyback transformer that was chosen has diodes in its internal construction, making it hard for AC excitation to perform well. An AC transformer is needed to utilize the full potential of this topology.

After the simulation of the ZVS topology, a DC approach was simulated. With this circuit, the controllability of the exciting circuit was the main objective. To do so a NE555 timer was used to generate a square wave signal, where its oscillating frequency can be controlled by a potentiometer. At its output, an LM741 Op-Amp was responsible for

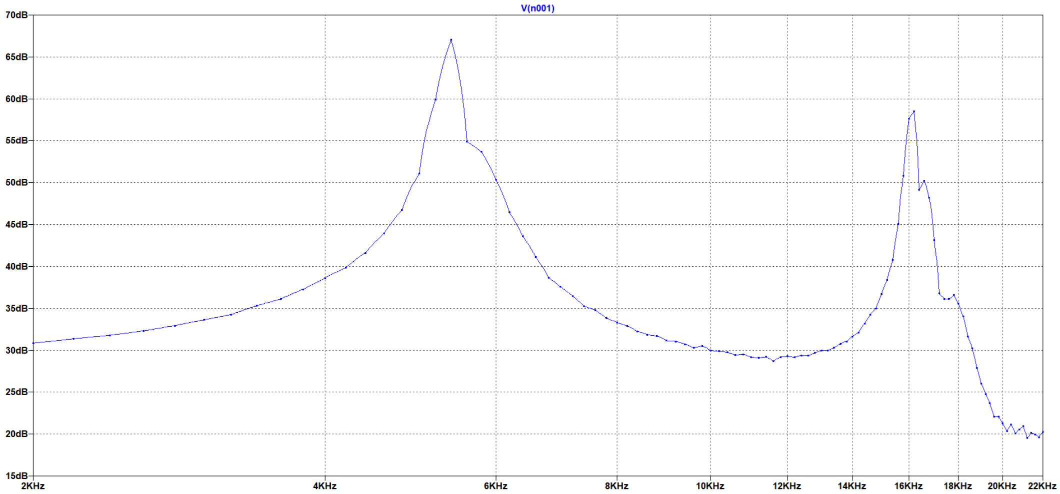

Figura 3. The Fourier power spectrum of the simulated signal at the output. The operating frequency is observed around 5 KHz, which is the resonant frequency for the components in the LC bank.

controlling, by means of a potentiometer, the width of the square wave signal driving the IRFZ44N power MOSFET. This can be seen in Fig. 4.

This controllability was key to understanding how to control the corona discharge parameters, reduce the occurrence of arcs, and control the input current and the thermal dissipation on the power MOSFET. As a future objective, the control potentiometers can be exchanged for a micro controller, allowing for a finer adjustment and reaction to the evolving conditions of the plasma. A PID algorithm can be inserted permitting active control of the discharge parameters.

3.2 Prototyped circuit

Aiming for simplicity, low-cost off-the-shelf parts, and lower input power requirements, the DC approach for plasma generation was selected. The circuit utilizes an NE555 timer responsible for generating a square wave, and then an LM741 Op Amp is responsible for exciting the NMOS IRFZ44N. A potentiometer connected to the feedback loop of the amp-op is responsible for Duty-Cycle control.

During testing a DC bench power-supply was regulated to 15 Volts and 2 Amps, where the circuit consumption was around 22 Watts. Lower voltages and input power were tested, including a 12 Volts input potential, confirming that it can be easily battery-driven.

In these experiments, the potentiometers were adjusted to a PWM signal with a frequency of 12 KHz and a duty-cycle between 40% and 60%. These ranges were adjusted to avoid electric arcs on the Petri dishes. Also, the distance between the positive electrode and the ground substrate was fixated at 50 mm. Inside the 5 mL disposable syringe, the electrode was at a distance of 15 mm from the exit point of the syringe.

With dimmed, or no ambient light, we can see a purple glow coming from the plasma wand. This glow is characteristic of atmospheric corona discharge, obtaining its purplish color from the ionization of atmospheric nitrogen. The corona discharge can be seen in Fig. 7

From the length of the arc, and taking into consideration air temperature, humidity, pressure, and the concentration of electric charges at the electrode tip and sharp edges, the output voltage was estimated to be around 6 kV and 8 kV.

4. CHARACTERIZATION OF BIOCIDE ACTION

Figure 9 demonstrates the sterilizing effect of the developed CAP source. The Petri dish on the left side is the control group, showing bacterial proliferation in the inoculated areas. Both the center and right side Petri dishes didn't present bacterial growth in the inoculated areas after being exposed to CAP for 10 minutes.

This test was done in duplicates, as seen in the center and right Petri dishes of Fig. 9. All inoculation points were bathed in 10 μL of 10^6 cells/mL. The specimen was *Peribacillus simplex* SF0016 (Orem *et al.*, 2019), in log phase, and the medium was LB with 1.2% agar, $40mL$ per Petri dish. After the exposure, all Petri dishes, including the control ones, were incubated for 24 hours at 30ºC.

During the test, a distance of 50 mm was set between the positive electrode, inside the disposable syringe, and the ground plane beneath the Petri dish. An electrical connection was made between the agar medium inside the Petri dish and the ground plane. This was necessary due to the DC nature of the discharge, which emits mostly one polarity species, causing an electrostatic charge to arise in the Petri dish, which repels the plasma stream. By grounding the dielectric material of the agar substrate and the ground plane, we can counteract this effect. The exit point of the serynge was

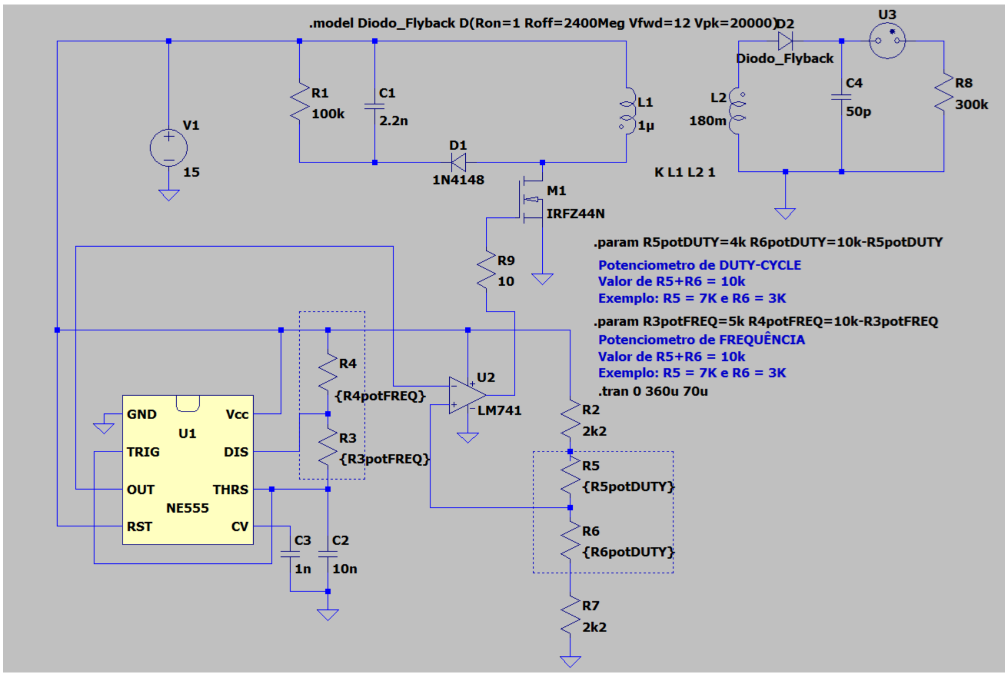

Figura 4. The DC circuit employed in the alpha version of the system.

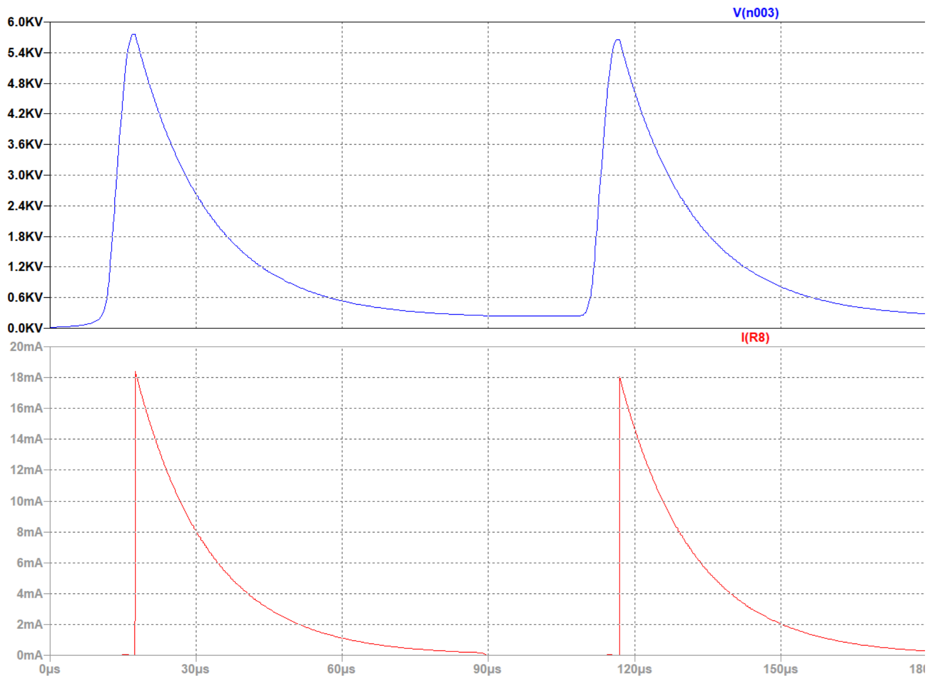

Figura 5. Output signal of the flyback transformer excited by a PWM signal.

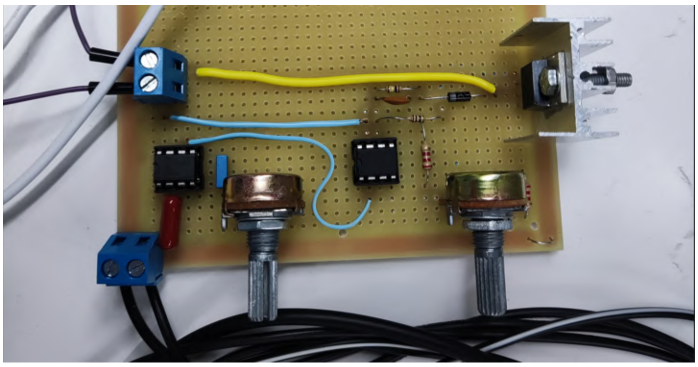
Figura 6. The circuit prototype for DC flyback activation.

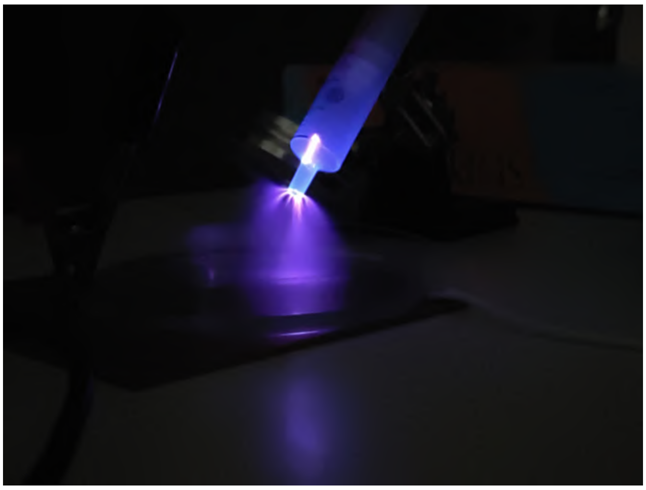

Figura 7. Corona discharge between the electrode inside the syringe and the ground plane.

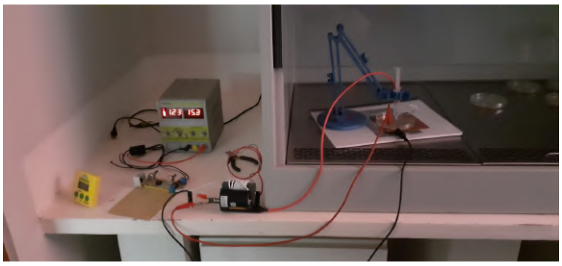

Figura 8. Experimental setup showing the prototype circuit and plasma application to a Petri dish for the characterization of the biocide action.

between 15mm and 20mm apart the surface of the agar substrate.

The prototype circuit performed well during this first test regime. However, arc discharges were observed during the tests. Arcs can occur due to the reduced dielectric impedance of the mixture of ionized and neutral species, the variance in distance between electrode and substrate, and environmental conditions, such as temperature, pressure, and humidity. When arc discharges indeed occur, either the NE555, LM741 ICs, or both can be catastrophically damaged. In the right conditions, especially with no arc discharges, it can operate for hours without flaws.

5. FINAL CONSIDERATIONS

In this work, a low-cost system capable of performing plasma sterilization has been developed. The list of materials employed to construct the prototype has a cost of less than $ 20. Preliminary tests demonstrate the effect of plasma on different biological contaminants. By comparing the results with untreated and uncontaminated plates, we have shown that the plasma source is able to sterilize inoculated Petri dishes.

Future work will aim to improve the CAP source. Some aspects to be improved are:

- The transformer excitation circuit is sensitive to electric arcs at the plasma wand output, burning several components throughout the tests.
- Some problems with electrostatic charging can be solved using AC circuits, such as the ZVS circuit, or by connecting the Petri dishes to circuit reference.
- Experiments with different exposure times are needed, to verify the minimum application time necessary and the relation between the input current and treatment time.

Currently, a new and improved circuit has been developed and is being tested to evaluate its reliability. In addition, a new type of electrode is in development alongside a new prototype with a broader application area.

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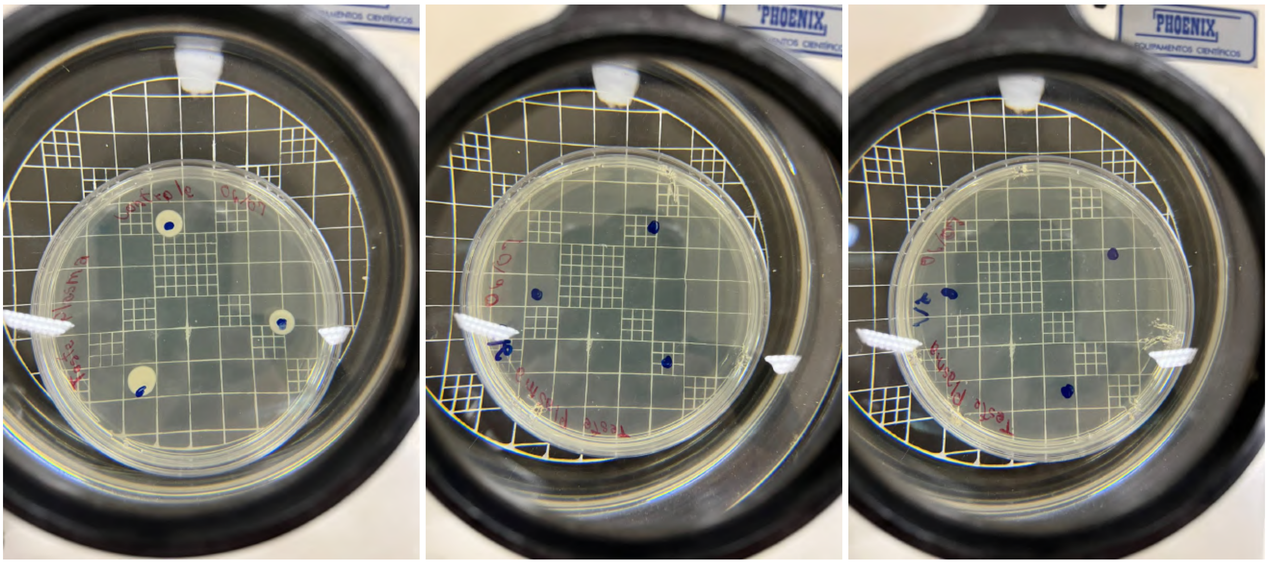

Figura 9. Preliminary results showing bacterial growth on the control Petri dish (on the left) and the duplicates that were exposed to 10 minutes of plasma treatment (center and right Petri dishes). All dishes were incubated for 24 hours at 30 ºC.

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