Development of a substrate-invariant 2-D array of nanosecond-pulsed streamer discharges

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Abstract
Two dimensional arrays of streamer discharges were developed using electrical discharge machining of stainless steel sheets and stacking them together with spacers to allow gas flow between the sheets. A nanosecond pulsing circuit, capable of delivering 2–40 kV pulses with pulse widths of >20 ns by using two spark gaps as switches, was developed as a simple tunable pulsing power supply. High resolution imaging of the plasma for uniformity across the array tips was conducted. Optical emission spectroscopy was used to characterize the species created as well as probe the temperature of the discharge for various substrates, voltage pulse durations, voltage pulse magnitudes, and gas flows. The discharge properties were found to be substrate independent for a wide variety of conditions.

1. Introduction

An array of tip electrodes is a relatively simple way to achieve wide-area treatment using pulsed discharges. They have a wide range of applications including sterilization [1, 2], air purification [3], surface processing [4, 5], swarm experiments [6, 7], and tumor treatment [8]. This paper seeks to extend the applications to include electroporation. Electroporation is the application of an electric field on a cell to induce pore formation in the cell membrane, introduce foreign matter into the cell, or destroy the cell membrane [9, 10]. New research has shown that nanosecond electroporation can also be used with some differing effects. Since the electric field is applied faster than the inherent charge time of ~100 ns for a cell, a higher electric field is possible before the charges migrate to reduce the applied electric field [11, 12]. Research shows that nanoparticles can be created using nanosecond-pulsed electric fields as well as other effects such as inducing apoptosis, damaging DNA, modifying caspase activity, and modifying calcium functions in the cell [12–14].

A challenge is generating sufficiently high electric fields which penetrate through the epidermis into the dermis, a distance of 68.9 μm or more [15], without penetration of the electrodes into the body. Applying higher fields nanoseconds in duration is one way to achieve the necessary high fields at depth, but inevitably this can create a plasma. In this work the generation of a plasma is taken advantage of in order to bring the potential of the electrode to the outer surface of the epidermis without direct contact. The constraints of this problem, namely high electric fields, nanosecond pulses, and no damage to the substrate, inherently lead one to investigate streamer and corona discharges. Corona discharges are typically weakly luminous, steady state discharges often where there is no conductive path to ground. The free electrons lose energy by collisions with neutrals until recombination occurs and the space charge is low enough to not greatly affect the overall electric field of the discharge gap. Streamers (sometimes called streamer–coronas) are growing ionized channels with significant space charge to affect the electric field that follow after the avalanche process of the primary electrons often caused in part by photoionization [16–19]. Once the distance between the electrodes has been traversed by a streamer the conductive channel grows forming a spark. If there is enough power to sustain the discharge it can become either a glow or an arc discharge depending on whether the discharge is sustained by secondary electron emission or thermionic emission, respectively. Thus streamer discharges are more preferred for electroporation applications over corona discharges but care must be taken that the streamer–to-spark transition does not occur.
The goal of this paper is to generate a short duration plasma with high applied voltage and field without damaging the substrate. Specifically the plasma uniformity, applied voltage, discharge temperature, and substrate compatibility are addressed.

2. Experimental setup

2.1. Array manufacturing

To achieve a high electric field, a high density of sharp tips was desired. This was achieved by electric discharge machining (EDM), where 0.0762 mm thick, 25.4 mm long sheets of stainless steel were stacked together and elliptical cuts were made along the length of one side of the stainless steel sheets to create an array of tips. Three ‘U’ shaped spacers were inserted between the EDM sheets to create a gas inlet, mixing chamber, and outlet near the tips by alternating their orientation, such that one opens up at the gas inlet and the other points in the opposite direction to be a gas outlet (see figure 1(a)). Spacers were also placed at the far ends of the array. The array of tips was then placed in a block of Teflon to hold them together and provide electrical insulation. A gas inlet was placed in the back of the Teflon block, and a high voltage wire was connected through the side to the stainless steel array. The final design is seen in figure 1(b).

2.2. RC nanosecond pulsing circuit development

A RC nanosecond pulsing circuit was developed which consists of a standard RC circuit ($R_1 = 10 \, \text{M} \Omega$, $C = 500 \, \text{pF}$) connected to a load by a spark gap switch (MSG). The load in parallel with the capacitor is a resistor ($R_2$), a second spark gap (SSG), and the electrode array (Load) in parallel with each other. The array is connected to the rest of the circuit via a meter-long twisted pair high voltage cable. A third spark gap (EXT) controlled by an external circuit (Amazing1 Ignitor10 power supply) was used for triggering the main spark gap switch (MSG). This triggering spark gap had to be placed sufficiently far away to avoid the high voltage side of the main spark gap from discharging to the ground electrode of the triggering spark gap rather than the load side of the circuit. As such, it was too far away to always trigger the main spark gap; jitter on the order of 1 microsecond was noted. The schematic of this RC nanosecond pulsing circuit is seen in figure 1(c). By varying the spark gap distances of the main switch spark gap (MSG) and the second spark gap (SSG), the desired pulse voltage and duration are able to be specified for a given load. The three types of loads tested were conductive, dielectric, and biological corresponding to a grounded metal electrode, a grounded saltwater bath with the glass petri dish acting as the dielectric, and pig skin thawed from frozen pork bought at the local grocer ~4 mm thick.

2.3. Uniformity investigation

Uniformity of the discharge across all of the tips was investigated using a Nikon D3300 camera with AF-S Micro Nikkor 105 mm lens. Initial testing was done using a fast ionization dynistor (FID) power supply (FID GmbH
model FPG 30-1NM, which produces 2 to 3 ns FWHM pulses. A parametric study was performed, with five gas flow conditions (air (4.3 SLPM), argon (3.61 SLPM), helium (8.54 SLPM), nitrogen (4.35 SLPM), and no gas all flowing into ambient air), four power supply conditions (1 kHz, 15 kV; 1 kHz, 30 kV; 10 kHz, 15 kV; 10 kHz, 30 kV), and three substrates (conductive, dielectric, and biological corresponding to metal, glass, and pig skin). Uniformity with the RC nanosecond pulsing circuit was also performed varying the three substrates, the peak pulse voltage, pulse duration, electrode-substrate gap, and air flow through the array. All images using the RC nanosecond pulsing circuit were taken at a pulsing frequency of 5 Hz (using BK Precision 4063 function generator to get the desired pulsing frequency). Oscilloscope traces were taken to measure the pulse magnitude and duration using a LeCroy WaveRunner 204MXi oscilloscope with North Star PVM-4 high voltage probe rated for up to 110 MHz bandwidth and a Bergoz current transformer with maximum rise time of 0.7 ns. Measurement locations are noted in figure 1(c).

2.4. Optical emission spectra and discharge temperatures
Optical emission spectra (OES) were collected for the RC nanosecond pulsing circuit for two wavelength ranges (352 to 358 nm and 375 to 381 nm) using an ICCD detector (Stanford Computer Optics 4Picos) attached to a 1 m focal length scanning monochromater (SPEX 1000 M spectrometer). The ICCD camera has a spectral resolution of ~0.04 nm, as measured by the FWHM of the iron 360.3 peak. The 2nd positive system of nitrogen along with the 1st negative system of N2+ was compared to modeled spectra from SpecAir [20] to determine the rotational (Trot) and vibrational (Tvib) temperatures of the nitrogen species in the gas. The discharge was pulsed at 5 Hz, and the emitted light was collected for ~3000 pulses (60 exposures each 10 s long). The parameters changed include the three substrates (conductive, dielectric, and biological), the pulse duration, the pulse voltage, the electrode-substrate gap, and the air flow rate. Example spectra with fits are seen in figures 2(a), (b) for both wavelength ranges.

Broadband OES were collected from 200 to 800 nm using a photomultiplier tube (PMT, Hamamatsu R928) for the dielectric substrate for five gas flow conditions (air (4.3 SLPM), argon (3.61 SLPM), helium (8.54 SLPM), nitrogen (4.35 SLPM), and no gas all flowing into ambient air) and four FID power supply conditions (1 kHz, 15 kV; 1 kHz, 30 kV; 10 kHz, 15 kV; 10 kHz, 30 kV) to determine the species generated. The PMT has a spectral resolution of ~0.2 nm, as measured by the FWHM of the helium 706.5 nm peak. OES were also collected for temperature determination of these conditions using three wavelength ranges (325 to 339 nm (Trot only), 347 to 359 nm (Trot and Tvib), and 363 to 382 nm (Trot and Tvib)). Example spectra with fits are seen in figures 2(c), (d), where the lower resolution compared to the ICCD detector can be seen, especially in resolving rotational lines. Spectra for the helium discharge was not modeled since the light levels for the 2nd positive system of nitrogen were too low for accurate modeling.
3. Results & discussion

3.1. FID testing

3.1.1. Uniformity investigation

Images were observed with varying the gas flow (figures 3(a)–(e)), power supply (figures 3(e)–(h)), and substrate (figure 4). Note for these images, there is reflection off of the bottom of the dielectric substrate, making the streamers appear longer than they actually are. Brighter discharges were noted at higher voltages and frequencies, as seen in the no flow condition in figure 3(e). Also, with higher voltages more of the array tips have visible streamers, showing a greater uniformity across the array. Helium created the most uniform discharge (figure 3(c)), having all of the tips on the array lighting up; all other gases (and no gas flow) were comparable in uniformity across the array. The conductive substrate generally led to brighter discharges compared to the biological and dielectric substrates, and the dielectric substrate generally had more uniform discharges across the tips. Uniformity on the biological substrate was difficult mainly due to non-uniformities in the pig skin itself (figure 4). The conductive substrate easily developed individual streamers that transition to a spark discharge. These are clearly seen in figure 4(b), where a few bright spark discharges are much brighter compared to the streamers from the rest of the array. The conductive substrate was also very difficult to align, thus the right side is brighter due to being slightly closer to the array.

3.1.2. OES using the FID

There was no observable change in the spectra other than increased light intensities for increasing voltage and frequency. The OES is seen in figure 5. In the spectra for both no gas flow and air (not shown) mostly only...
nitrogen (2nd positive system) was observed with a few trace impurities. In nitrogen, NO (gamma system) was also observed. For argon, OH (A-X system) dominated the spectra, with some nitrogen (2nd positive system) and argon visible. Trace amounts of NO (gamma system), O (777 nm), and H (656 nm) were noted in helium along with various helium peaks. OH (A-X system) and N2+ (1st negative system) dominated the helium spectra. Due to high ionization energies for argon and helium, excited species are able to greatly increase chemical reactions for other species. Unlabeled peaks are second order peaks from the diffraction grating.

From modeling the temperatures, air (and no gas flow with discharge into ambient air) had the highest vibrational temperature and lowest rotational temperature (figure 6). Argon had the highest rotational temperature and the lowest vibrational temperature; however, this analysis may not be accurate since it does not account for excitation of the nitrogen species from collisions with argon metastable species. In general higher vibrational temperatures are correlated to higher electron temperatures which correspond to higher E/n, thus it is likely that air has a higher electric field. This is supported by the fact that air is electronegative, making it harder to break down and requiring a higher E/n. There was no large trend in the spectra for higher voltages or frequencies. An IR thermometer was used to measure the substrate temperature after testing but no significant temperature rise (<5 °C) was noted. This is not unexpected since the plasma pulse duration is quite small compared to the overall treatment time.

3.2. Nanosecond pulsing circuit
3.2.1. Circuit characterization
The pulses from the RC nanosecond pulsing circuit were very repeatable, with a peak voltage variability under 5%. Characteristic voltage and current pulses are seen in figure 7 for two different breakdown voltages of the main spark gap (MSG). Figures 7(a), (b) shows a lower voltage case where the peak voltages and currents are fairly similar for both the metal and dielectric loads. The energy per pulse is approximately 9 mJ for the conductive load and 12 mJ for the dielectric load, where most of the energy is delivered in the initial pulse. For the lower voltage cases (for a 5 mm electrode-substrate gap it was below 18 kV and 30 kV for the conductive and dielectric substrates, respectively) the air gap acts as a dielectric and voltage reflection can occur, leading to pulse voltages higher than the breakdown voltage on the capacitor. For the higher voltage cases, such as in figures 7(c), (d), the metal and dielectric loads have a larger variation in peak voltages and voltage reflection does not occur.
There is also a significantly higher peak current which was not able to be measured. For the largest voltages (>30 kV) and capacitances tested (5 nF) the peak instantaneous currents are on the order of 1 kA.

Pulse frequencies from a single pulse to ∼100 Hz can be achieved depending on the current supplied from the power supply (the power supply was limited to 7.8 mA). The pulse voltage has a minimum value around 2 kV, due to issues in reliably spacing the main triggering spark gap. Peak pulse voltages were generally close (within about 3 kV) to the breakdown voltage on the capacitor (slightly higher if voltage reflection occurred). There was a roughly 1 kV ns⁻¹ rise and fall on the voltage pulse if the second spark gap was set below a threshold.

Figure 6. N₂ 2nd positive system temperatures for dielectric substrate using FID power supply.

Figure 7. Voltage and current traces from the RC nanosecond pulsing circuit for a 5 mm electrode-substrate gap for a MSG breakdown voltage of (a), (b) 9.0 kV and (c), (d) 27.5 kV for the (a), (c) conductive substrate and the (b), (d) dielectric substrate. Plasma energy is shown in black in (a), (b).
value; larger second spark gap distances did not reliably discharge, leading to longer pulse durations with slower fall times, and shorter values reduced the peak pulse voltage but still maintained the 1 kV ns\(^{-1}\) rise and fall speeds.

### 3.2.2. Uniformity investigation

Overall three main discharge modes were noticed with the RC nanosecond pulsing circuit. The first mode is streamers mainly only visible on the edge of the array directed toward the substrate, which occurred for all three substrates, seen in figure 8(a) for the biological substrate, figure 8(c) for the conductive substrate, and figure 8(e) for the dielectric substrate. Figure 8(b) shows a different angle for the biological substrate to more clearly distinguish that there is no visible discharge in the center of the array. This is likely due to the outer tips having a slightly greater negative differential resistance, leading them to preferentially break down. Since the FID power supply has a faster voltage rise time, this effect was not observed with that power supply. This is the most common mode observed, occurring for all variations with pulse duration. This first mode was also observed for all pulse voltage variations for both the biological and dielectric substrates. When the voltage was increased above a threshold or the electrode-substrate gap reduced below a threshold for the conductive substrate the second discharge mode was observed as seen in figure 8(d). The discharge would begin like the first discharge mode, but then one or two of the streamers from individual tips would become a preferential path for current and thus grow much brighter as that streamer begins transitioning to a full spark discharge (note that figure 8(d) is actually 25 distinct discharge events, thus multiple discharges are seen). The third discharge mode was observed for the dielectric substrate with no gap between the array and the substrate as seen in figure 8(f), where the streamers spread out across the substrate, instead of being directed toward the substrate. This occurs because the surface becomes charged, thus the streamers must spread out further to find ground.

### 3.2.3. Temperatures

Overall for all substrates the rotational temperatures are low, suggesting the discharge is not transitioning to a glow mode \([21]\) but remains as a streamer discharge (figure 9). This occurs because the voltage pulse is too short for the discharge to transition to another mode, irrespective of the substrate used. However, the temperature of the second discharge mode seen in figure 8(d) was not able to be measured due to the presence of iron peaks in the nitrogen spectra, which indicate electrode ablation. This low rotational discharge temperature holds true (figure 10) for varying the applied voltage (i.e. maximum peak pulse voltage) from 12.4 kV to 26.3 kV, varying the pulse duration (FWHM) from 15.9 ns to 33.8 ns, varying the air flow rate from 0 SLPM to 8 SLPM, and varying the electrode-substrate gap from 4 mm to 9 mm.

It was noticed that the 378 nm peak family consistently showed lower temperatures (both \(T_{rot}\) and \(T_{vib}\)) than the 356 nm family. This could be due to the effect of N\(_2^+\) concentration on the modeled spectra in the 356 nm family, while there are no N\(_2^+\) lines in the wavelength range for the 378 nm family. Overall, the temperatures were \(T_{rot}=310 \pm 35\) K and \(T_{vib}=4208 \pm 717\) K. Some rotational temperatures were observed to be lower than ambient. This is due to some error in the modeling; however it was consistent and should not affect any trends visible in the data.
4. Conclusion

An array was successfully developed to maximize the electric field and allow a gas to flow through it during operation. No large changes were observed in varying the array geometry with regard to tip spacing. Initial FID testing suggests air to have the highest electric field of all gasses tested due to its electronegativity, making it the most attractive for an electroporation application. A RC nanosecond pulsing circuit was developed to power the array electrode, capable of producing pulses from 2 to 40 kV with pulse durations ranging from a single pulse to $10^{-100}$ Hz depending on the voltage and current supplied from an external DC power supply. The discharges from the array using the RC nanosecond pulsing circuit appear very uniform over a wide range of conditions and substrates, except that the center of the array does not commonly light up. Over a large range of conditions and over various substrates the discharge temperature is highly non-equilibrium, with rotational temperatures near ambient.
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