Electrode Erosion in an Atmospheric Pressure Nanosecond-Pulsed Non-Thermal Plasma Source

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Abstract: Electrode erosion was investigated in a pin-to-pin nanosecond-pulsed plasma source operating in argon at atmospheric pressure. Scanning electron microscopy (SEM) and optical emission spectroscopy (OES) were used to observe morphological changes of the electrode tips and to correlate such changes to metal vapor emission lines. Results indicated that erosion is occurring in both the diffuse and spark regimes and that electrode tip erosion affects run-to-run repeatability.

Keywords: Pin-to-pin discharges, nanosecond-pulsed discharges, electrode erosion.

1. Introduction

Non-equilibrium atmospheric pressure plasma sources have been developed for an array of novel applications including gas conversion, treatment of liquids, and surface functionalization[1]. Many of these technologies aim to generate a diffuse non-thermal plasma volume consistently. Though advantageous, reproducing such a volume with the same set of electrodes poses unique challenges, frequently encountered during the development phase of novel plasma technologies. For instance, a given plasma system can no longer re-establish a diffuse plasma volume after a few successful runs under identical operating conditions. Existing literature suggests that the challenges associated with reproducing diffuse plasma volumes are the result of the shape and surface condition of the electrodes gradually changing, especially after exposure to the spark regime[2], [3]. Although suspected, the effects of electrode erosion on non-thermal plasma systems remain mostly unexplored. In this work, we aim to establish that electrode erosion is occurring in a pin-to-pin non-thermal plasma source and that it can be detected using a non-invasive diagnostic technique. The overall goal of this work is to gain a quantitative understanding of the possible implications of electrode erosion on a nanosecond-pulsed, non-thermal plasma discharge source operating at atmospheric pressure.

2. Methodology

A pin-to-pin electrode configuration was powered by a repetitive nanosecond-pulsed power supply sustaining non-thermal plasma discharges in argon at atmospheric pressure (Praxair, 5.0 Ultra High Purity). The pin electrodes were made from stainless steel 316 rods (0.16 cm OD), which were manually sharpened and polished to an angle of 15° using a 3D-printed sharpening guide. The tip-to-tip distance between electrodes was maintained at 10 mm. A polycarbonate tube (1.9 cm ID, 45 cm long) was used as a plasma chamber. Argon was injected at 10 SLPM using a single port and exited through four drilled holes downstream from the electrodes, at the end of the polycarbonate tube. System pressurization was prevented by monitoring the pressure gauge. The polycarbonate tube was capped by a 3D-printed viewport which contains a quartz window to facilitate the collection of spectroscopic data. Figure 1(a) illustrates the field of view of the spectrometer through the viewport. A homemade dual-polarity high-voltage nanosecond-pulsed power circuit capable of producing total peak voltage drops ranging from 5 to 15 kV was used to power both electrodes at a frequency of 1 kHz. The total peak voltage drops depended on the desired plasma regime being generated. Voltage measurements were gathered using a 1000x high-voltage probe (Tektronix, P6015A, 75 MHz bandwidth). The discharge current was measured using a current monitor positioned just before the cathode (Pearson Electronics, 500 A, Model 6585).

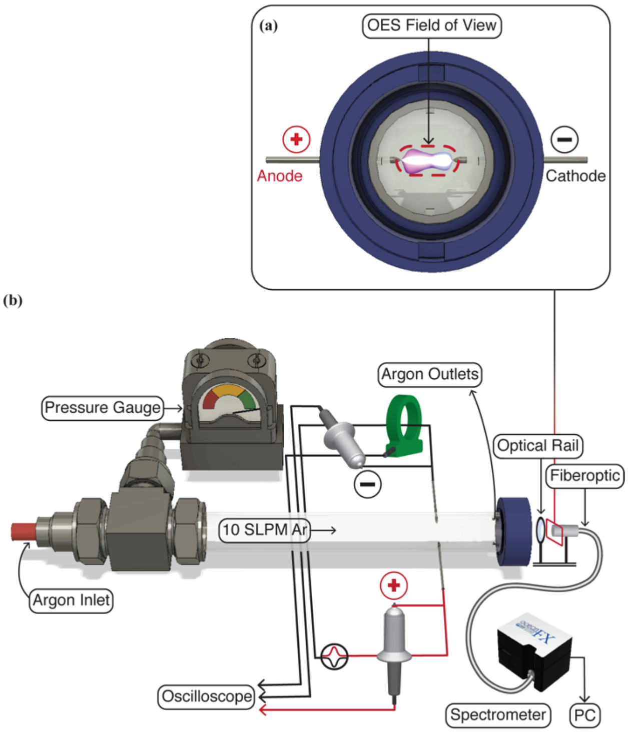

Figure 1. Experimental configuration **(a)** OES field of view and **(b)** side view.

Optical emission spectroscopy (OES) and scanning electron microscopy (SEM) were utilized to identify and quantify erosion effects. Optical emission spectra were recorded using an Ocean FX spectrometer (Ocean Optics, 25 µm slit, extended range 200-1100 nm) with an optical resolution of 0.78 nm. The optical rail was configured to allow the spectrometer to collect all light emitted by the plasma discharge occurring between the electrodes. The light collection area of the spectrometer is highlighted in red in Figure 1(a). Integration times varied based on the emission intensity of the discharge. Hence, the OES data are both spatially and temporally integrated. Scanning electron microscopy (SEM) images were collected using a variable-pressure environmental scanning electron microscope (Thermo Scientific Quanta, F50 ESEM) at 15 kV with a 3.0 (r.u.) spot size under high vacuum conditions. The experimental methodology of this work followed two phases. The objective of the first phase was to gain insight as to how electrode erosion can be detected and quantified using OES and SEM techniques. So, the spark regime was studied first, as it was hypothesized that this was the most likely condition under which evidence of electrode tip erosion could be detected. The strongest atomic iron emission lines were identified and reasoned to represent electrode erosion occurring in the plasma volume. The second phase of the experimental methodology required the application of this information to the diffuse regime to search for erosion effects.

3. Results and Discussion

We counted 49 emission lines over the observable spectral window from the spark regime (14.4 kV), found in Figure 2. The spectrometer was set to 0.5 sec integration time and averaged ten scans. Three voltage conditions were included to illustrate the evolution of the plasma discharge towards the spark regime.

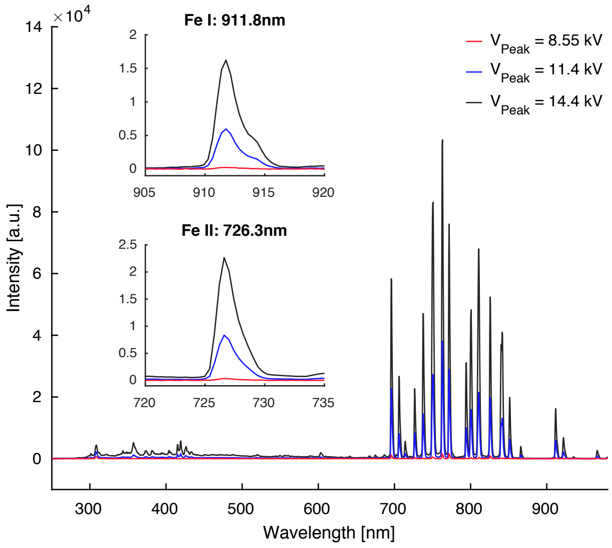

Figure 2. Emission spectrum of the discharge in the spark regime.

All peaks were identified using the NIST atomic spectra database and corroborated where possible using literature references reporting on similar plasma systems[4], [5]. This experimental phase yielded valuable information regarding which atomic transition lines were indicative of erosion. Since the stainless steel 316 electrodes are predominantly composed of iron, many spectral peaks were identified as atomic iron transitions. The emission spectra also contain several emission lines from argon in the 700-900 nm range, and a significant OH emission band, which appears as a line due to low spectrometer resolution, around 308 nm. We associate the OH emission band to the presence of 3 ppm of H_2O in the argon tank, and not ambient air, as it appears consistently throughout all experiments and with similar intensity values.

Emission lines of interest were selected to create a group of transitions that were representative of the sublimation of iron from the electrodes. The transitions were selected based on emission line intensity, frequency of appearance, and excited species type. Thus, the 49 emission lines were narrowed down to four main iron transitions, considered as indicators of the sublimation of iron from the electrodes. The excited species of interest are summarized in Table 1, where E_k represents the upper energy levels for each transition. Atomic transition lines at 921.8 nm and 415.0 nm are reported as blended lines, so the identified species are reported as multiple excited states of iron[4].

Table 1. Spectral lines of interest.

Species	**λ [nm]**	**E_k [eV]**	**Intensity [a.u.]**
Fe I, Fe II	921.8	14.10	8.5×10^3
Fe I	911.8	4.19	2.0×10^4
Fe II	726.3	14.05	2.1×10^4
Fe I, Fe II	415.0	9.07	6.9×10^3

In addition to optical evidence of erosion, SEM images of the cathode before and after spark regime experiments were collected to corroborate suspected erosion effects, illustrated in Figure 3. A distinct change is visible in the surface morphology and radius of curvature of the cathode. Particular attention should be paid to the changing radius of curvature, as it has been demonstrated to limit the existence of the glow regime at atmospheric pressure[2], [3]. In this context, a parameter limiting the existence of the glow regime at atmospheric pressure would directly influence run-to-run repeatability.

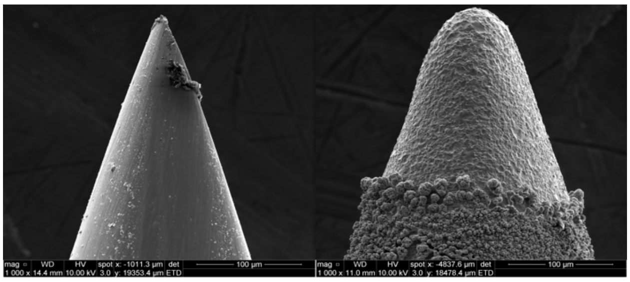

Figure 3. SEM images of the cathode tip before (left) and after (right) exposure to the spark regime.

The second phase of the work applied the information gained from the spark discharge experiments to search for evidence of erosion in the diffuse regime. Figure 4 presents the spectral data acquired from a diffuse non-thermal plasma discharge established at three voltage conditions, where the spectrometer integration time was set to 2.0 sec. The integration time had to be increased because the overall emission intensity of the discharge was significantly lower than in the spark regime. Figure 4 also illustrates a notably decreased signal-to-noise ratio relative to Figure 2. Attempts to improve the signal-to-noise ratio were made by acquiring frequent dark spectra with the plasma off, which were later subtracted from the bright signal acquired with the plasma on, in addition to increasing the integration time. Although somewhat effective, the decreased signal-to-noise ratio evident in Figure 4 resulted in the inability to detect and isolate lower intensity emission lines for analysis. This limitation presented an additional challenge with regards to determining which iron emission lines could be captured by the spectrometer in both the diffuse glow and spark regimes. Glow-to-spark experiments were conducted with continuous OES measurements to narrow down which iron emission lines were the best indicators of electrode erosion.

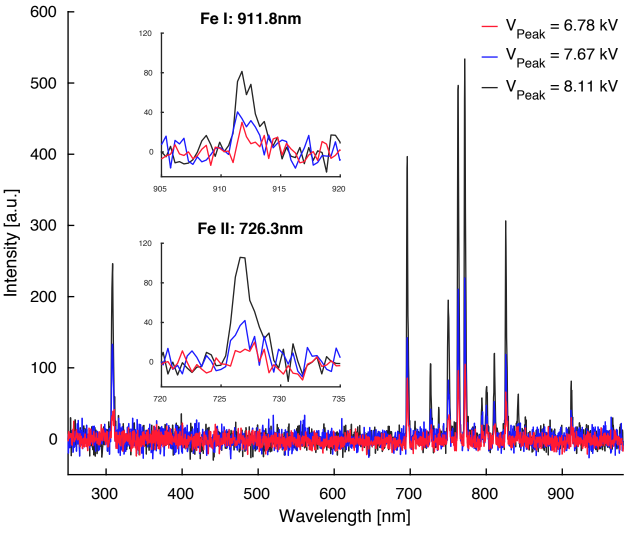

Figure 4. Emission spectrum of the discharge in the diffuse regime.

Analysis of the glow-to-spark experimental data involved calculating the area under the peak of interest (emission line intensity) and plotting the area as a function of increasing applied voltage starting in the diffuse glow regime, all the way to the spark regime. This method clarified which iron emission lines were visible in both the diffuse, transition, and spark regimes: 911.8 and 726.3 nm. Figure 5 shows how the emission intensity from the 911.8 nm line (Fe I) changes as a function of applied voltage. Each experiment was conducted with a new set of sharpened electrodes. The error introduced from realigning the electrodes between experiments, in addition to slight differences in sharpness, results in the slight variations in the curves seen in Figure 5. However, the curves generally follow a similar trend as the system transitions from a diffuse discharge to the spark regime.

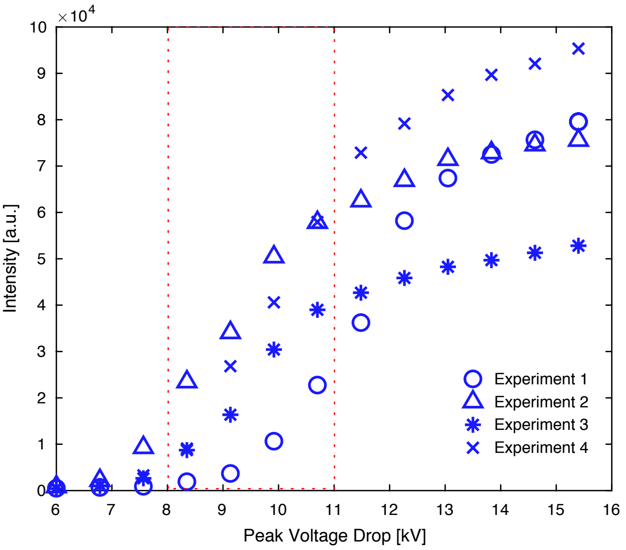

Figure 5. Emission intensity (a.u.) of the 911.8 nm (Fe I) atomic line as a function of the total voltage across the pin-to-pin electrode gap.

The evolution of the emission intensity from the 726.3 nm line (Fe II) as a function of applied voltage was also analyzed and is shown in Figure 6. The same slight differences between experiments are apparent, following similar trends as Figure 5. Interestingly, all curves seem to follow a roughly sigmoidal trend as the plasma transitions from a diffuse glow to a spark. The peak voltage drop range from 8 to 11 kV highlighted by a dashed line contour in both Figure 5 and Figure 6 indicates when the diffuse glow transitions to spark. In this range, the rate of change of the erosion curves increases rapidly. Once the pin-to-pin voltage exceeds 11 kV, the rate of change of the curves decreases, seeming to stabilize, except for experiment 1, which stabilizes around 13 kV. Identifying the transition to spark from the spectroscopic data allows for further investigation of the possibility of preventing transitions to spark in non-thermal plasma systems, thereby prolonging electrode lifetimes.

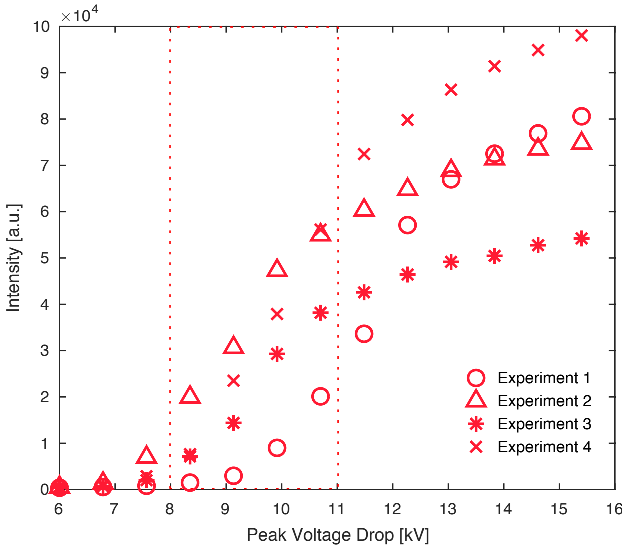

Figure 6. Emission intensity (a.u.) of the 726.3 nm (Fe II) atomic line as a function of the total voltage across the pin-to-pin electrode gap.

Considering the intensity values from the 911.8 nm and 726.3 nm emission lines as representative of erosion, their abruptly increasing slopes indicate that iron sublimation accelerates once the system begins to transition to the spark regime between 8 and 11 kV. Electrode erosion under these conditions could be increasing the radius of curvature of the electrode tips, which in turn makes it more difficult to re-strike a plasma using the same electrodes. Interestingly, the small electrode degradation revealed by the increased radius of curvature of the electrode tip significantly alters its ability to strike a non-thermal plasma at atmospheric pressure.

4. Conclusion

Using a pin-to-pin electrode configuration to sustain repetitive nanosecond-pulsed atmospheric pressure non-thermal plasma discharges in flowing argon, we investigated the possible effects of electrode erosion on re-ignition characteristics using optical emission spectroscopy (OES), scanning electron microscopy (SEM) of the electrode tips, and electrical diagnostics.

The first phase of experiments utilized the spark regime to gain insight as to how erosion could be quantified in the system. We identified the 911.8 nm (Fe I) and 726.3 nm (Fe II) emission lines as indicators of the sublimation of iron from the electrode tips. Glow-to-spark experiments indicated that electrode erosion is detectable in all plasma regimes. Erosion effects were demonstrated to accelerate via emission line intensity data once the system begins to transition to spark between 8 and 11 kV. We suspect that the electrode erosion occurring in the diffuse glow regime increases the radius of curvature of the electrode tips and thereby causes issues with run-to-run repeatability. The long-term goal of this work is to leverage the data gathered from this experimental configuration to develop an online tool allowing researchers to mitigate electrode erosion effects, using a non-invasive technique like optical emission spectroscopy. Achieving this goal will require future investigation into the relationship between current and radius of curvature of the electrodes.

5. Acknowledgements

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6. References

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