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Intense and Highly Energetic Atmospheric Pressure Plasma Jet Arrays

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INTENSE AND HIGHLY ENERGETIC ATMOSPHERIC PRESSURE PLASMA JET ARRAYS

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ABSTRACT

This thesis documents the efforts taken to produce highly ionized and concentrated atmospheric pressure plasma using an arrayed atmospheric pressure plasma jet (APPJ) system. The honeycomb-shaped array features seven plasma jets operating in close enough proximity to one another to exhibit jet-to-jet coupling behavior. Optimal gas flow rates for the system were determined and intense plasma plumes composed of argon and/or helium are generated. Optical emission spectroscopy was employed to observe the charged particles responsible for the emissions of each gas discharge and APPJ operation mode. Plasma etching of indium tin oxide glass was conducted to verify the highly energetic properties of a plasma generated using two dissimilar gases, in order to confirm the possibility of plasma coupling between them.

ACKNOWLEDGMENTS

I would like to thank Dr. Sung-O Kim for the research opportunity and access to his lab and resources, without which this thesis would not have been possible. COMSET provided excellent laboratory and office space. I owe much thanks to Dr. Jae Young Kim for his endless support and advice. His expertise in the field of atmospheric pressure plasma was invaluable, and without his help and guidance, I doubt I would have been able to survive in this field. To the other members of Dr. Kim's group during this work, Jae Young Kim and Do Yeob Kim, thank you for being friendly and supportive and helping to alleviate the tedium of work. To family and friends, thank you for all of your encouragement, and for tolerating my odd behavior and erratic schedule. I am glad to have had such amazing help and support from everyone mentioned, you all made this possible.

Mon professeur n'a pas le moindre sens de l'empathie. Éviter cet homme à tout prix.

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CHAPTER ONE

INTRODUCTION

Nonequilibrium plasmas, possessing relatively high electron temperature and low gas temperature, are becoming very widely studied due to their useful characteristics. Such plasmas exist at a much lower temperature and higher pressure than thermodynamic equilibrium plasmas while maintaining many beneficial properties such as enhanced plasma chemistry and generation of high concentrations of reactive species [1]. Atmospheric pressure plasma, as it is referred to in this work, can exist in open air, at relatively high pressure and any atmospheric composition. Additionally, it exists at relatively low temperatures, far lower than those required to sustain plasma ionization. Such cold plasmas have a very low rate of ionization, typically around 2%. Consequently, they have a very low charged particle density, and very little power is required to generate this kind of plasma [2]. Numerous devices have been developed to generate atmospheric pressure plasma, but one of the most promising is the plasma jet.

An atmospheric pressure plasma jet (APPJ) offers many advantages for application over other plasma systems. Arguably the most significant property it possesses is its relative safety. An APPJ generates low (<100°C) temperature plasma, which can be made appropriate for use on living tissue or thermally sensitive substrates. For the type of device used in this study, the gas is excited by an electric field, and there is minimal risk of electric shock or generation of ozone. The plasma can be sustained in open air, removing the need for expensive vacuum equipment or environmental control.

Construction and maintenance of atmospheric pressure plasma systems is less complicated and labor-intensive than low pressure, high temperature systems. For applications requiring plasma application on a small area or with precision, an APPJ is preferable over dielectric barrier discharge plasma devices [3]. Finally, these systems can use inexpensive gases and be constructed from cheap materials while still delivering stable and consistent plasma discharges. With these advantages, the atmospheric pressure plasma jet is sure to find use in many industrial and experimental processes.

Currently, atmospheric pressure plasma is being used for numerous applications. There is an ever-growing body of research related to biomedical applications of plasma. It can be used to kill cancer cells or microorganisms while leaving healthy tissue intact, or quickly induce blood coagulation [3]. Atmospheric pressure plasma systems can also be used to sterilize items, etch surfaces, deposit compounds, or decontaminate food items in industrial processes [2]. One shortcoming of these systems is that their low energy compared to vacuum plasma precludes them from many potential applications. For atmospheric pressure plasma, it is difficult to excite electrons enough that they ionize neutral gas species. This is a consequence of the high population of gas species present in high pressure environments which reduces the mean free-path for electrons. Extensive studies and effort have been invested in increasing the energy contained in cold plasmas to increase their efficacy and usefulness.

The basis of this thesis is increasing the energy in atmospheric pressure plasma using an array. The intention is to utilize jet-to-jet coupling to produce intense and highly energetic plasma. By operating multiple plasma jets in close proximity to each other, the plasma energy can be increased and focused into a single plume. This plume exhibits higher energy than a single jet acting alone and accordingly is observed to generate more reactive species [4].

A device was constructed to observe the effects of varying gas flow rates on the jet-to-jet coupling behavior. A honeycomb-structured 7-array plasma jet was designed such that the central tube and six outer tubes operate on separate gas flow systems. High purity (99.999%) He and Ar gases were used to generate atmospheric pressure plasma and the optical intensity and optical emission spectra were recorded for varying plasma generation conditions. Previous studies involving larger plasma jet array structures have demonstrated that more intense plasma discharge is possible with modest increases in power consumption. In the case of a 19-tube array, it was possible to double the plasma energy of a single tube plasma jet while consuming only 20% more power [5]. The procedures described in later sections are intended to determine the optimum conditions for generating intense plasma with the seven tube array, as well as identify the accompanying changes in plasma composition. The purpose of the following experiments is to increase the energy contained in atmospheric pressure plasmas, enabling their use in more applications.

CHAPTER TWO

THEORETICAL BACKGROUND

2.1 Atmospheric Pressure Plasma

By definition, plasma is a state of matter similar to a gas, in which a portion of the particles are ionized. Artificial plasmas have been researched extensively in order to take advantage of many of the unique and useful properties of plasmas. The attractive properties of plasma include high electrical conductivity, high chemical reactivity, and release of electromagnetic radiation. Artificial plasmas do not exist in thermodynamic equilibrium, their electron and ion temperatures differ. If the energy of an ion or electron is too low, a collision will simply result in the recombination of the particles, and the pair returning to a bound state as a gas. As a result, nonequilibrium plasmas require constant excitation (usually in the form of an applied electric field or heat) in order to remain ionized.

A gas is much easier to ignite into a plasma when it exists at a very low pressure (<10mTorr), generally referred to as vacuum plasma. In the case of low pressure, the gas density is relatively low, and thus the mean free path for electrons is fairly large, resulting in a greater acceleration of electrons within the gas during excitation from an outside source. These electrons create high energy collisions which cause the molecules in the gas to ionize and transition between quantum states. For such plasma, the temperature tends to be very large due to the high energy state of most of the particles contained in it. Vacuum plasma has some inherent drawbacks such as the need for expensive vacuum equipment and difficulty of direct application, which has led to the study of atmospheric pressure plasma.

In the case of atmospheric pressure plasma, the degree of ionization tends to be very low, typically only 2% of the gas. Even so, it will still exhibit the same useful properties of a more highly ionized plasma discharge. Compared to vacuum plasma, it is generated at high (relatively) pressure, in open air (760 Torr). In this type of discharge, excitation of the bulk gas results in a lower proportion of ionized particles due to the lower energy of particle collisions (mean free path). This also results in a lower plasma temperature, which can be a strong advantage when using this type of plasma. Unfortunately, the main disadvantages of this type of plasma stems from its low rate of ionization. In this work, efforts are taken to improve the energy contained in atmospheric pressure plasma using the jet-to-jet coupling effect of an atmospheric pressure plasma jet array.

An atmospheric pressure plasma jet (APPJ) is a plasma device which creates a plasma plume in open air. The construction of this experiment's device uses a single electrode to energize the plasma, using an ITO glass plate as a ground. The plasma plume may also be sustained in open air, using the surrounding air as a virtual ground if the excitation voltage is high enough to produce a stable plasma discharge. The plasma generated by these devices remains at or near ambient temperature, and poses little risk of electric shock while boasting high stability. Due to its relative safety, atmospheric pressure plasma is ideal for use in sensitive processes or on living tissue, as seen in Figure 2.1. As discussed in later sections, multiple plasma plumes generated near each other can interact and merge if the conditions are right. This phenomenon results in a more concentrated and energetic plume of plasma, which is of great value to most applications it can be used for. Additionally, as seen later in the plasma etching tests, some types of plasma are typically precluded from some applications due to their low energy, but the intense plasma generated by the coupling behavior can make them more viable [1].

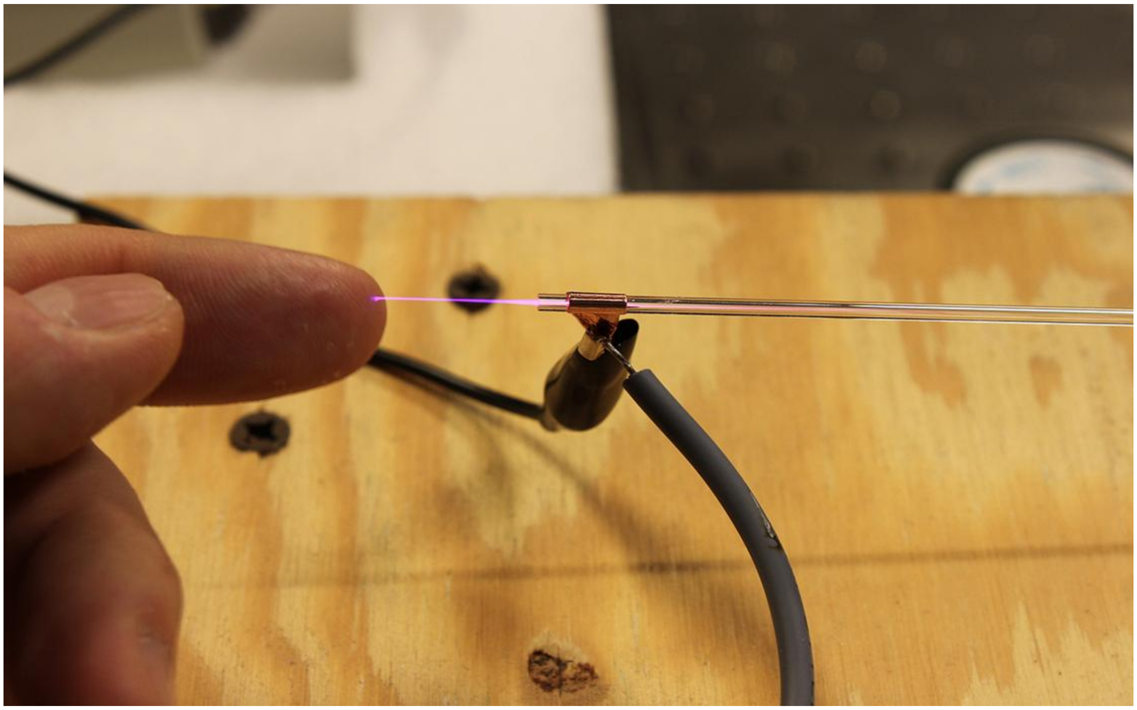

Figure 2.1: An atmospheric pressure plasma jet.

2.2 Plasma Spectroscopy

An atom or ion will emit radiation when transitioning between various quantum states. Many different types of radiation can occur in plasma. Line radiation occurs for electron transitions between bound levels, creating line spectra. Other forms of radiation such as recombination, free-free (Bremsstrahlung), and even Cherenkov radiation either do not occur or are very rare at the temperatures and energy levels of low temperature plasma [6]. When plasma is being generated in open air, the charged particles have the opportunity to react with atmospheric gases. These interactions lead to the creation of reactive species, which are of great interest to most of the applications of such a plasma system. Within this study, a spectrometer measuring the band of light between roughly 180 and 900 nm wavelength is used. With reference to a database of atomic and molecular spectra, it is possible to identify some of the reactive species being generated in plasma.

For the argon and helium plasmas generated for the experiments contained in this thesis, the line radiation resulting from excitation of atmospheric diatomic nitrogen is a common element of the plasma emission spectrum. A nitrogen molecule can be transferred from its ground state $N_2(X^1\Sigma_g^+)$ to an excited state $N_2(C^3\Pi_u)$ via collision with electrons with an energy greater than 11.0 eV. Subsequently, the excited nitrogen molecule transfers to the $N_2(B^3\Pi_g)$ state by emitting a photon of 337.1nm wavelength. If the energy in an electron exceeds 18.7 eV, nitrogen ions $N_2^+(B^2\Sigma_u^+)$ will be produced instead. These particles emit photons of wavelength 391.4nm when transferring to the $N_2^+(X^2\Sigma_g^+)$ state. The incidence of light emissions at either aforementioned wavelength is indicative of these reactive species in plasma [4, 7]. Similarly, other peaks present in plasma spectral data correspond to specific charged particles, and can be used to easily identify the generation of known reactive species.

2.3 Plasma Physics

The phenomenon of interest in this work is the jet-to-jet coupling behavior exhibited by plasma discharges in proximity with one another. The primary cause of this behavior is the electrical coupling of charged particles. Additionally, the presence of a common ground electrode allows the charged particles to merge and form a common discharge path [4]. Although ionized particles will interact with each other, high degrees of gas turbulence can prevent the merging of charged particles.

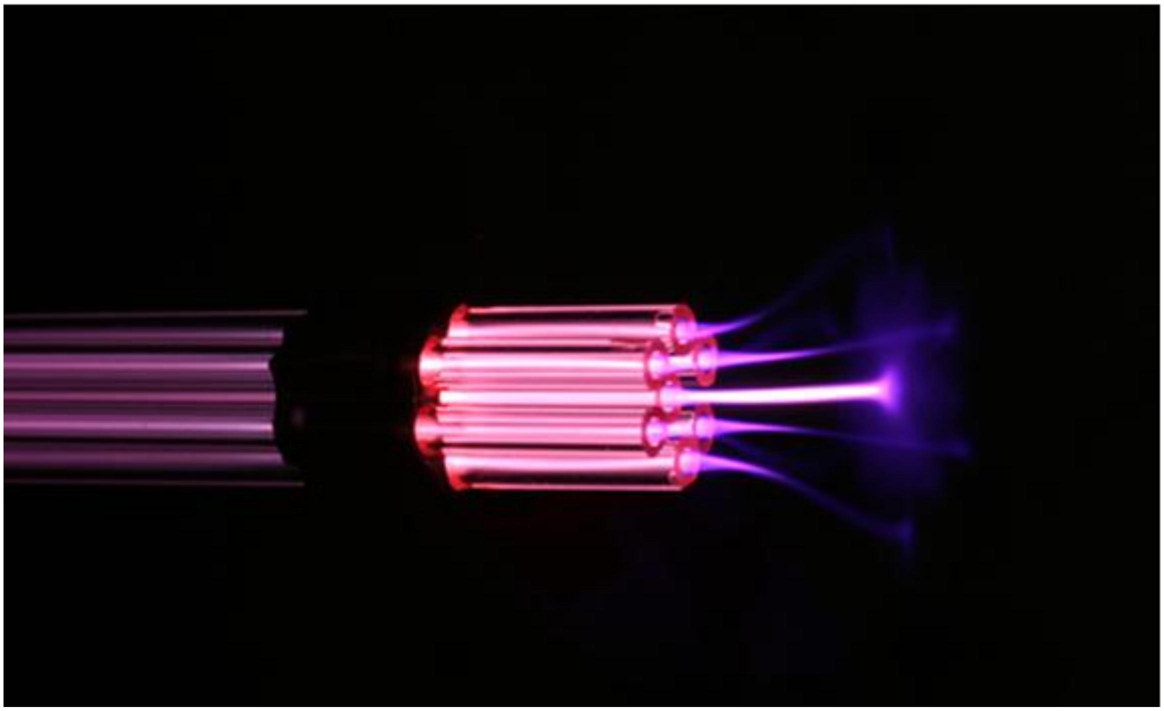

Figure 2.1: The jet-to-jet coupling behavior in effect.

Figure 2.2 illustrates the jet-to-jet coupling phenomenon. The APPJ array is seen contacting an ITO glass ground plane (not visible due to darkness). Each outer plasma plume can be seen losing intensity before contacting the ground plane. Most of the ions originating from the outer tubes have merged into the central plume, the remainder of unabsorbed ions form filaments which stretch towards the ground electrode in different positions. The central plasma discharge is noticeably more intense than the surrounding plumes. In the course of these experiments, this behavior is used to generate the most concentrated plasma possible in the central plume using a jet array device.

The phenomenon of jet-to-jet coupling is believed to be due primarily to the plasma behavior of filamentation (Birkeland currents). Highly ionized plasmas exhibit a distinctive behavior in which large current densities within the plasma are condensed into string-like structures. This happens because of the electromagnetic fields projected by the collections of charged particles contained in plasma. The plasma thus self-constricts into thin strands of energetic matter, which can be best described and modeled as a magnetic fluid. Within atmospheric pressure plasma discharges, all of the requisite components for this behavior to occur are present, and the cohesion of singular plasma plumes as well as the merging of multiple plumes is evidence of this type of activity. Additionally, once created, the central plasma channel appearing as a filament boasts a significantly increased electrical conductivity relative to the surrounding environment, and will draw additional electrons and ions into itself, thus maintaining its intensity.

With an atmospheric pressure plasma jet, an additional factor of gas turbulence is introduced into the plasma. It has been observed in previous experiments [1] that the flow rate of gas in a plasma jet may be too great for the capacitively coupled plasma discharge to occur between the plasma jet electrode and the ground plane. When this occurs, the plasma channel between these areas cannot form and the typically intense and resilient plasma channel ceases to form entirely. The severity of such turbulence is so great that even the virtual ground of surrounding air cannot be used to produce displacement current sufficient enough to sustain plasma. Additionally, it has been observed that sufficiently high gas velocities can overpower the attractive forces between charge particles and force multiple adjacent plasma discharges to remain separate. Within the following procedures, care must be taken to ensure that the flow of feed gas does not interfere with the desired plasma coupling behavior.

2.4 Plasma Etching

The primary mechanics of plasma etching, discussed later in this work, are the sputtering effect of ion bombardment and chemical interaction of reactive species at the target surface. The reactive species in plasma may interact chemically with the surface of a target material and alter its composition or remove atoms. The efficacy of this action is generally dependent on the reactivity of the specific free radical species generated in plasma. Ion bombardment is more often the primary agent of etching in cold plasmas. When an ion strikes the surface of a material, it sets off a series of collision cascades within the material. When these cascades recoil and return to the surface of the material, an atom may be ejected if the cascade energy exceeds the surface binding energy. For atmospheric pressure helium plasma, an ionized helium particle possesses relatively little mass and not highly energetic. Thus, the energy imparted to a material during collisions is low, and unlikely to result in the ejection of atoms from a material surface [3]. Coupled with the low chemical reactivity of the glass used in these experiments, atmospheric pressure helium plasma typically makes a poor etching agent. Figure 2.3 illustrates the action of etching using a plasma jet.

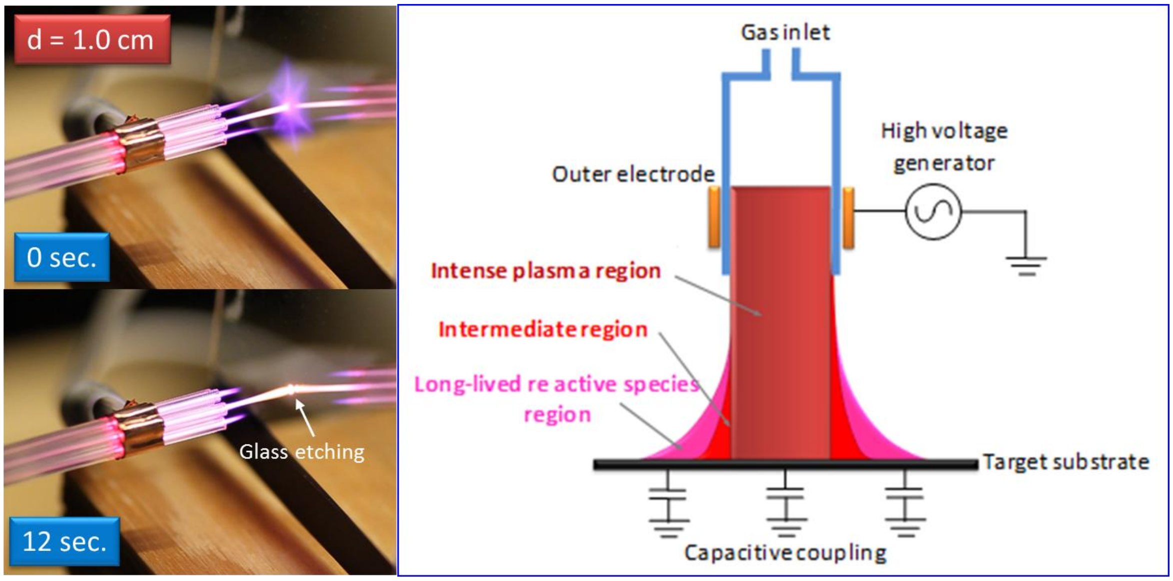

Figure 2.3: Etching with an atmospheric pressure plasma jet.

The above figure shows the different regions of a capacitively coupled plasma discharge and the difference in behavior when etching begins. The primary activity of the plume exists in the intense region, directly between the jet aperture and the point of contact on the ground plane. During non-etching operation, the discharge will splay outwards across the surface of the glass plane, creating the long-lived reactive species region. When etching begins, the reactive species and ions that would normally be ejected outwards by the gas flow are instead acting in the intense region at the point of contact on the plane. This activity is reinforced and intensified by the decreasing thickness of the target substrate. The reduced thickness will reduce the resistance and capacitance of the material at that point, and cause the plasma channel to flow selectively through that area until it is moved a sufficient distance away. As seen in later experimental results, the etching of a surface may be used as an indication of the energy contained in plasma.

CHAPTER THREE

EXPERIMENTAL PROCEDURES

3.1 Device Fabrication

The Atmospheric Pressure Plasma Jet array device was constructed to allow an isolated gas flow in the central tube, and a uniform flow rate amongst the six outer tubes. The seven tubes consist of 2mm outer diameter, 1mm inner diameter quartz glass tubing, bound together in a honeycomb-patterned structure. The tubes are surrounded by a plastic case serving as the gas chamber for the outer tubes. The plastic case is connected to a gas flow via a flexible plastic tube. The central quartz tube is isolated from the outer tubes and extends through the case into a steel tube connected to a separate gas system. The seven tubes are bound 1 cm from the end of the cluster by a copper tape electrode. All gaps and junctions in the gas system are made airtight using a vacuum-rated epoxy resin. The result is a plasma jet which can change operation modes based on the gas flow rates in the central and outer tubes. The jet also operates without direct electrode contact, and produces its plasma via capacitive coupling between the jet's bound copper electrode and the target, which acts as a ground.

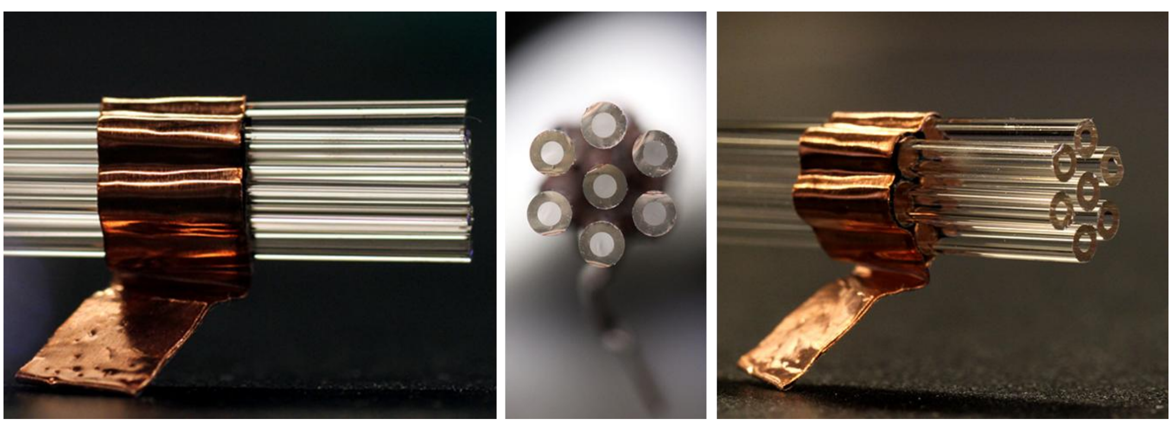

Figure 3.1: The atmospheric pressure plasma jet array: side, front, and perspective views.

3.2 Driving System

The gas flow in the plasma jet is ionized indirectly by a high voltage source. The driving circuit provides tens of kilovolts at tens of kilohertz. The typical driving conditions for helium and argon-based plasma in these experiments are 15 kV p-p at 35 kHz in open air. Once a plasma plume is created and stabilized, it is placed into contact with the non-conducting side of an ITO (indium tin oxide) glass plate of 0.8 mm thickness. The ground electrode is attached to the conducting side of the glass (not in contact with the plasma). In all experiments, the tip of the plasma jet is placed 1cm from the ITO glass plane. Because the plasma used in these experiments is not excited by direct contact with an electrode, the current flow and power consumption are very low.

3.3 Optical Measurement

The optical intensity of the plasma plume is captured via a Hamamatsu C6386-01 photo sensor amplifier. The amplifier converts light to voltage, and connects to the digital oscilloscope to provide a signal corresponding to the light intensity of the plasma in real time. The instrument's sensing lens is placed opposite the central plasma plume's point of contact on the glass side of the transparent ITO plate. This photo sensor connects directly to the digital oscilloscope and its optical intensity output can be observed alongside the other measured waveforms. Any changes in the experiment parameters such as gas flow or supply voltage generate instantaneous results in the oscilloscope. Using this system, maxima for peaks in optical intensity were determined via observation and gradual change in gas flow rates. The luminosity data from the photo sensor is not measured in any specific unit. Throughout these experiments, the instrument can be assumed to be operating on the same scale for any given set of plots and its results are intended only for comparison of data sets belonging to the same test. The optical emissions measured for the plasma detailed in this study occur between 300nm and 800nm. The photo sensor amplifier used to measure optical intensity is logarithmically more sensitive to increasing wavelength within the bounds of these experiments' parameters. Figure 3.3 contains the device characteristics.

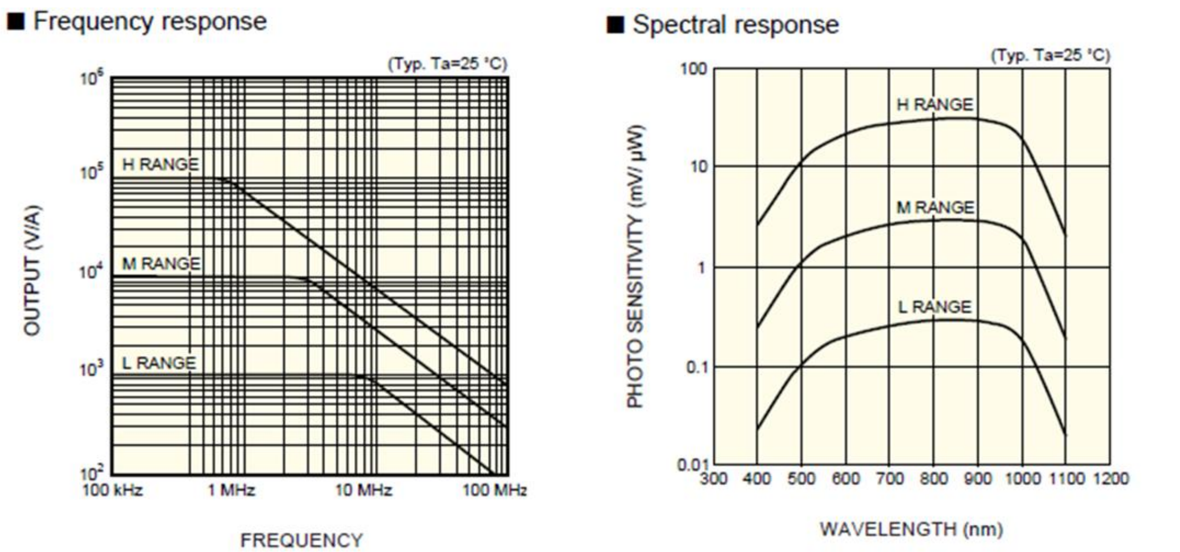

Figure 3.2: Response characteristics of the photo sensor amplifier.

3.4 Spectral Data

The reactive species generated by an atmospheric pressure plasma jet are of great interest and use to the applications involving these devices. The different wavelengths of light composing the optical discharge from plasma were measured using an optical emission spectrometer (Ocean Optics USB-4000 UV-VIS) connected to a computer. Each unique peak observed in the light spectrum corresponds to a charged particle. The relative prevalence of known reactive species and ions can be determined by referencing these results with known atomic and molecular spectra. The optical emissions for pure hydrogen plasma are already known and well-documented, as well as those for argon [8]. In a later part of the experiments detailed here, plasma emission composed of both gases is observed, and analysis of the optical emission is critical for explaining the plasma interaction. For the spectrometer used, the units of luminous intensity are arbitrary, and the device operates at the same sensitivity for all gathered data. The only exception to this is a set of data gathered through a neutral density filter in order to bring some peaks into the device's range of sensitivity.

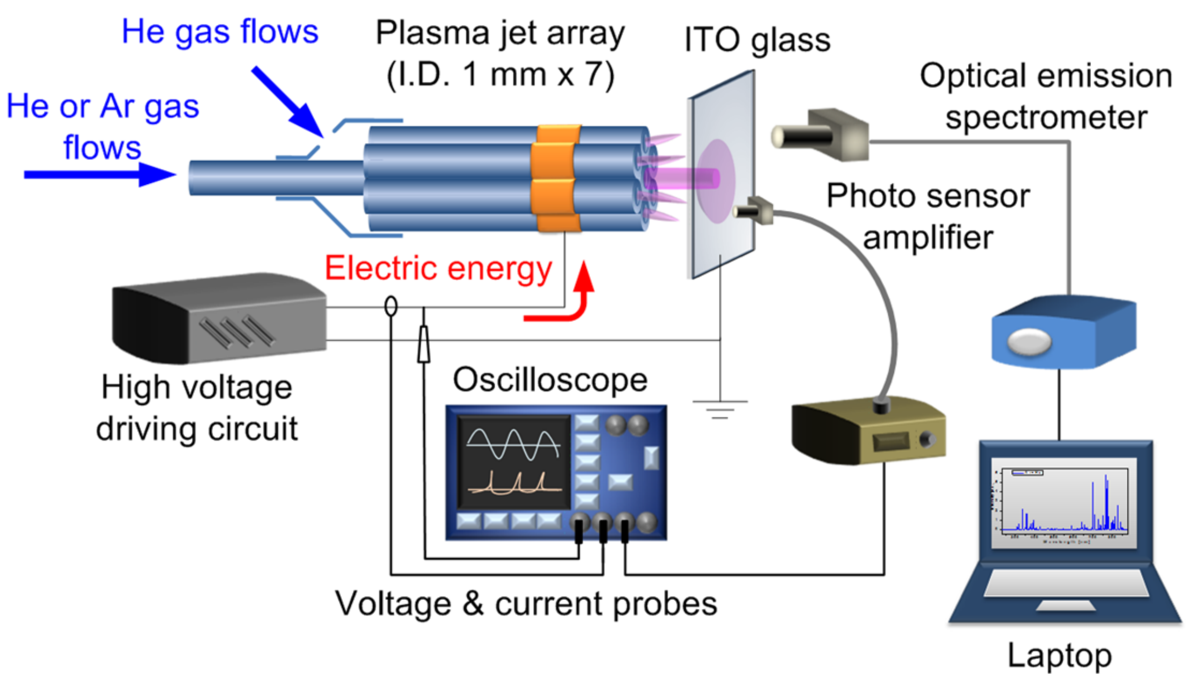

Figure 3.3: Complete atmospheric pressure plasma system.

CHAPTER FOUR

DATA ANALYSIS AND RESULTS

4.1 Two Discharge Modes in APPJ

The atmospheric pressure plasma jet array fabricated for these experiments is intended to operate at the maximum possible intensity for a device of this size. The variable uniform gas flow in the outer tubes allows for fine-tuning of the availability of supplemental charged particles in order to produce a central plasma plume of optimal energy.

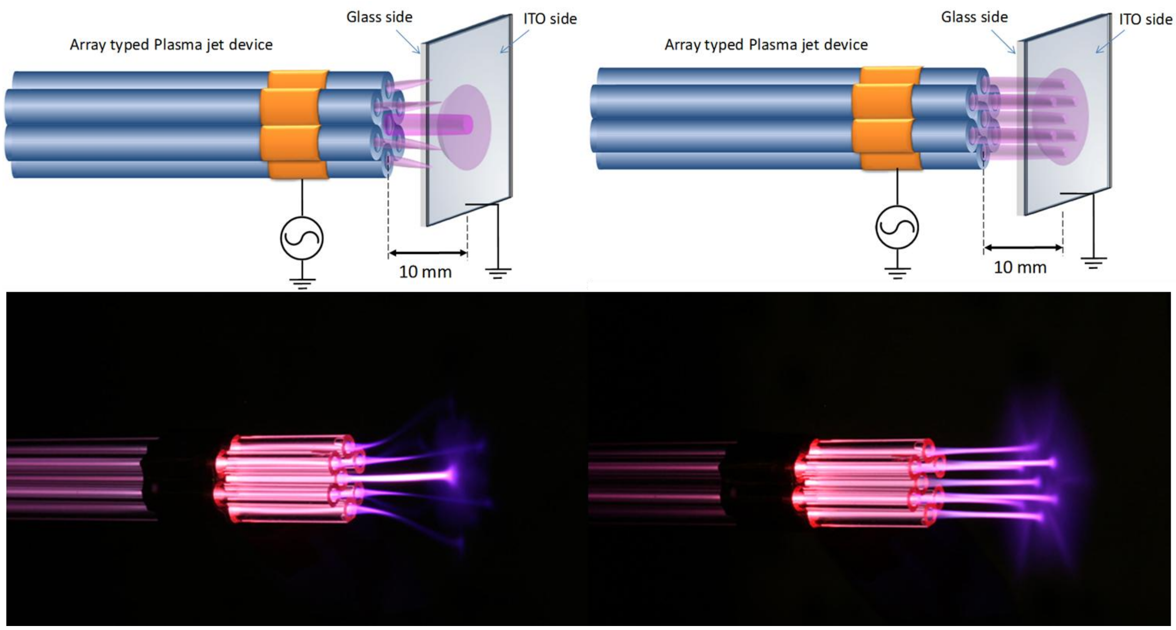

Figure 4.1: APPJ array displaying intense (left) and well-collimated (right) modes of operation.

For an array of plasma jets operating very close to each other, charged particle interactions can greatly influence the shape and behavior of their plasma plumes. When several smaller gas flows are excited near a plasma plume, they will donate ions to the plume and increase its intensity. This type of activity is visible in the left panels of Figure 4.1. If the surrounding gas flows are too fast, the turbulence will hinder plume interaction and cause each tube to act more independently of its neighbors as seen in the right panels of Figure 4.1. When the outer plasma plumes are contributing to the central plume, the APPJ array is said to be operating in intense mode. When each tube produces a plume that contacts the ground plane in a separate location with minimal interaction between flows, it is operating in well-collimated mode. The optical intensity of the central plume of such an array operating in well-collimated mode is comparable to that of a single jet acting alone.

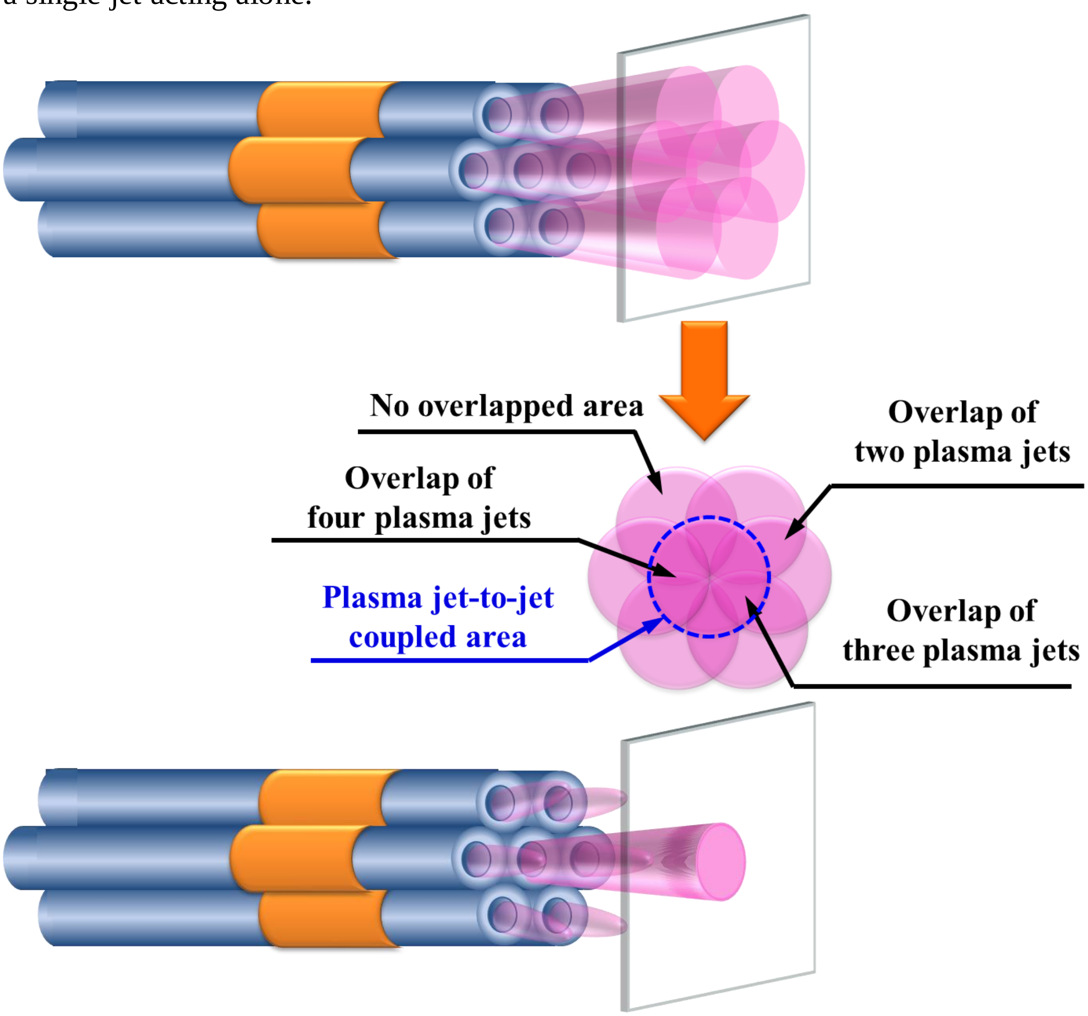

Figure 4.2: The jet-to-jet coupling effect due to plume overlap.

Figure 4.2 demonstrates how multiple adjacent plasma plumes can overlap to form an intense central plume. The discharge from each glass tube has a wider diameter than the tube itself. When multiple tubes are bound closely together, as with this array structure, several regions of overlap are created. The greatest region of overlapping plumes appears in the center of the array, and the effect is a significant increase in the intensity of the central plume while the outer plumes are diminished. The limiting behavior of the jet-to-jet coupling effect was explored in the following experiments.

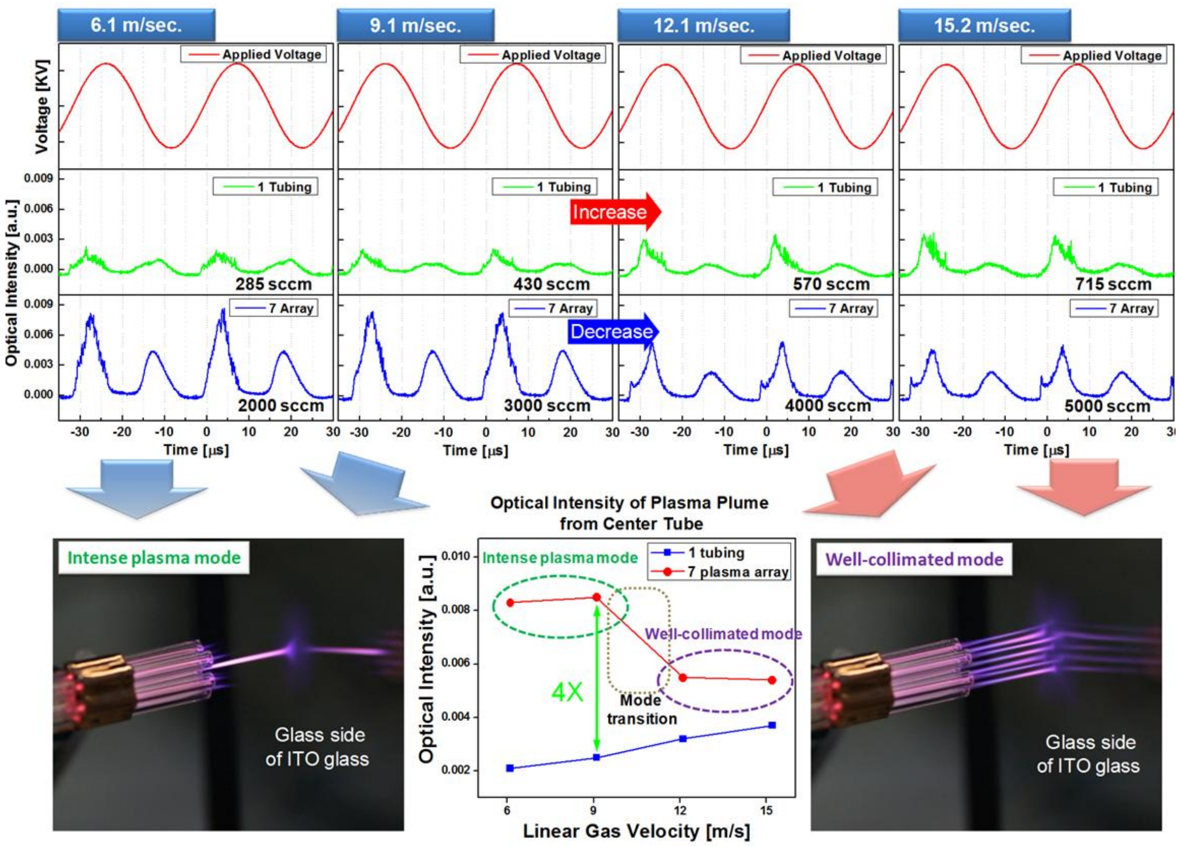

Figure 4.3: Plasma jet behavior for uniform flow rate in each tube.

Figure 4.3 illustrates the activity of a plasma jet with equivalent gas velocities in each tube. As the total gas flow rate increased, the velocity in any given tube must increase as well. For lower speeds (9.1m/s and 6.1m/s), the surrounding tubes contributed charged particles to the central plume. At gas velocities exceeding 12.1 m/s, the outer plumes contacted the ITO glass plate and decreased the intensity of the central plume. This manner of operation is referred to as well-collimated mode [4]. Compared to a single jet, the array operating in intense mode was approximately 4x more intense and energetic. This data shows that an array of this size with uniform flow rates can produce intense plasma with a significant improvement over single jets.

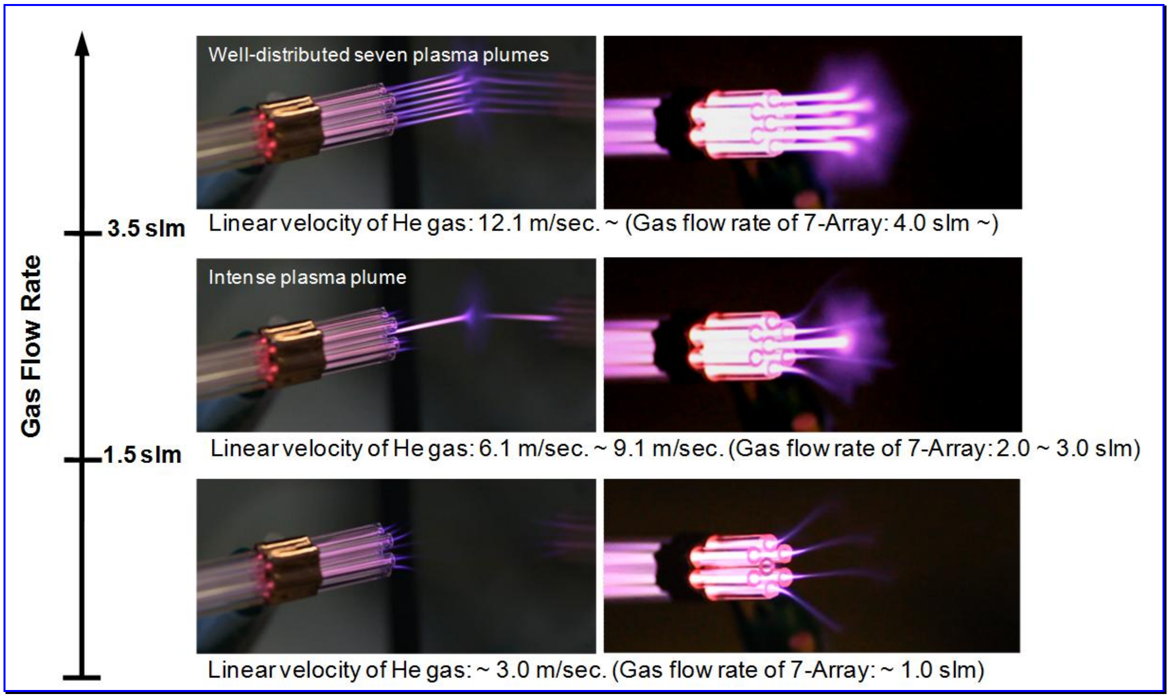

Figure 4.4: The behavior of the plasma jet array with variable total gas flow rate.

Over the course of these experiments, it was discovered that the optimal conditions for intense plasma discharge were primarily based on total gas flow rate. The best results in any case resulted for a distance of about 10mm from the ground plane. Increased distance prevented the plasma plume from forming between the jet and ground plane. A distance of less than 10mm resulted in lower central plume stability and in increased prevalence of well-collimated behavior for this device. Additionally, the optimal flow rate for this device was found to be between 1500 and 3500 sccm total gas flow. A deficiency of gas would prevent the intense plasma plume from forming, while a surplus would generate only well-collimated behavior. These results indicate that for this single electrode plasma jet, there is a peak in intensity of the central plume for a specific set of constraints.

4.2 Intense Atmospheric Pressure Helium Plasma

In order to determine the ideal gas flow rate mixture for helium plasma in this device, a series of tests was run. For these tests, the central gas flow was increased in increments of 400sccm. For each increment, the single jet, maximum intense, and maximum well-collimated optical intensities were recorded. As seen in Figure 4.3, the plasma was most energetic at a total flow rate of approximately 2640sccm. Until turbulence due to excessive gas velocities renders the plasma plumes unable to interact with each other, the primary factor in helium plasma intensity appears to be total helium flow. Additionally, the phenomenon of an increasing intensity during the falling slope of the excitation becomes prominent at higher total flow rates. This behavior may produce larger peaks of activity, but its effect on the average emission is minor due to its relatively short duration. In Figure 4.5, the most energetic plasma was produced with 1200sccm flowing in the central tube, with 240sccm per outer tube. These helium flow rates correspond to gas velocities of 25m/s and 5.2m/s respectively. These numbers imply that producing the most intense plasma with this device is dependent upon the center tube having a large flow rate supplemented by small, non-disruptive gas flows.

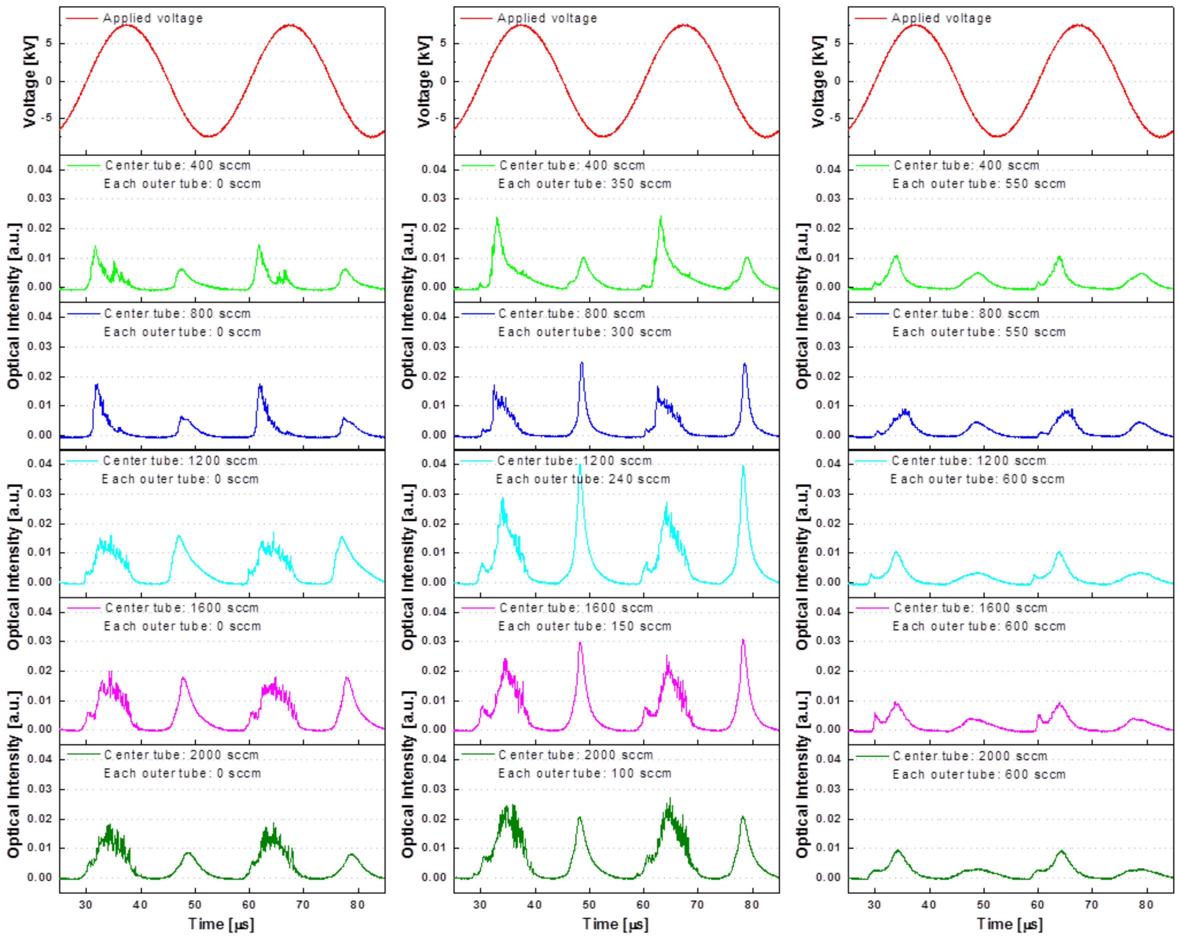

Figure 4.5: Optical intensity of He plasma driven at 15kV p-p and 35kHz.

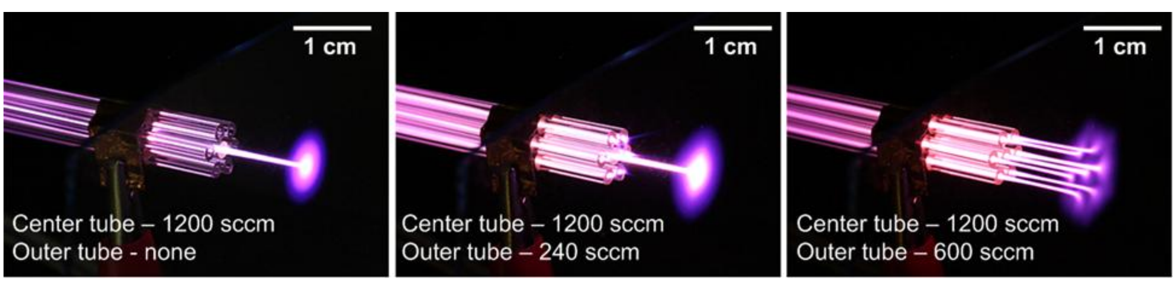

Figure 4.6: Three operation modes of the APPJ. From left to right: single jet, intense, and well-collimated modes.

Figure 4.6 illustrates the modes of operation for the APPJ array. The behavior of the outer tubes is of great interest in optimizing plasma emissions. For a maximally intense plasma plume, the outer tubes must have enough gas flowing to provide a significant amount of charged particles without causing turbulence. In the middle image, the outer tubes can be seen to have small plumes beginning to form, but not contacting the ground plane.

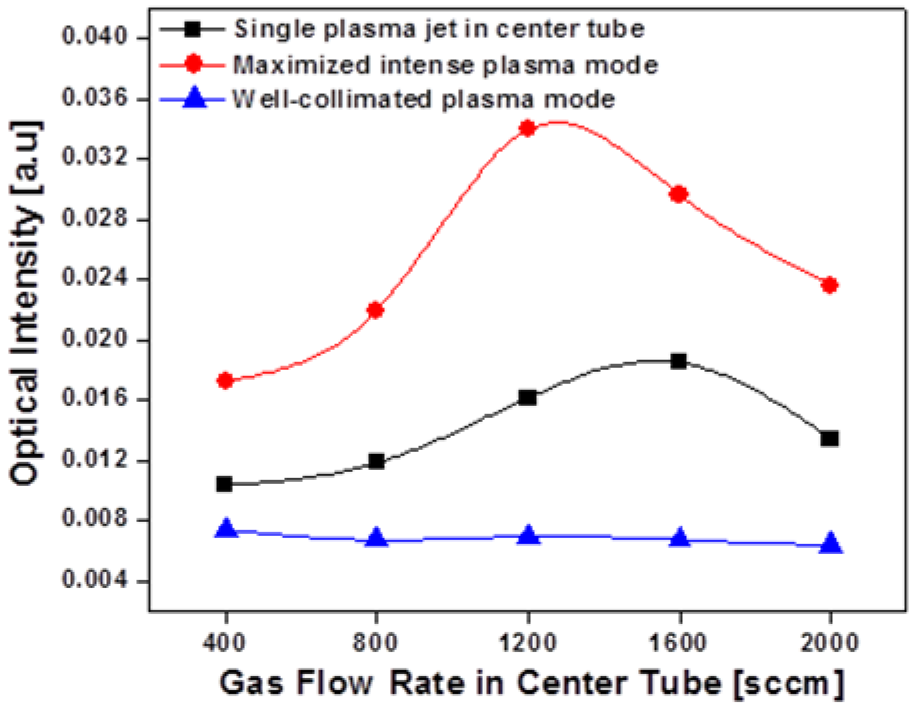

Figure 4.7: Maximum intensity for single tube, intense and well-collimated plasma modes.

Figure 4.7 shows trend lines for the maximum possible optical intensity of helium plasma based on central flow rate. The points corresponding to 1200sccm and 600sccm were chosen for optical emission spectroscopy. For each of these values, the intense and well-collimated modes were observed in order to determine the relative quantity of reactive species generated. Figure 4.8 displays the OES results for the intense plasma formed with 1200sccm as the center flow.

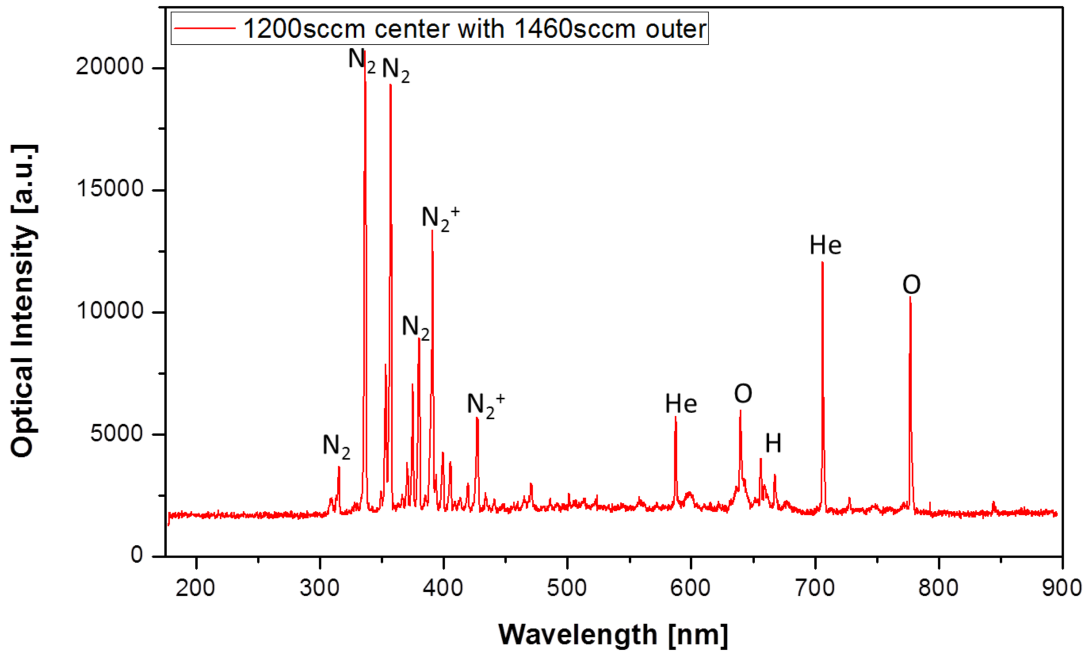

Figure 4.8: Optical emission spectrum of intense helium plasma. Each notable major peak has been labeled with the particle it corresponds to.

The spectral data for the most intense incidence of all-helium plasma (1200sccm + 1460sccm) displays primarily the existence of excited N_2, N_2^+, He, H, and O in distinct peaks. The presence of O and H atomic spectra in the results is due to the generation of this plasma in open air. For this type of plasma discharge, diatomic nitrogen is the primary influence on the optical intensity of the plume. Additional OES data for the other flow rate combinations chosen for analysis is available in Appendix A.

4.3 Intense Atmospheric Pressure Argon-Helium Plasma

With the properties of all-helium plasma studied, the focus of the experiments shifted to the combination of argon and helium in a similar assortment of tests. Argon requires a greater applied voltage and/or gas flow rate than helium in order to provide a similar glow discharge [2]. For tests utilizing argon as the central gas flow with the same excitation, it can be expected that the plume will reach its maximum intensity with a greater gas velocity than that required for helium. The argon plasma discharge was supplemented with helium gas flows in the six outer tubes. The primary interest in pursuing this plasma mixture was optimizing one plasma discharge by coupling the charged particles from a less intense plasma into it. In the case of pure helium plasma, the most common reactive species emitting radiation in the visible spectrum are ionized He, N_2, and N_2^+. For argon plasma, the molecular nitrogen peaks remain, but are much less significant compared to the intensity of the ionized argon spectral emissions [8]. Despite the large difference in intensities of Ar and He ionization emissions, it is a point of interest to quantify the potential for jet-to-jet coupling between two different gases and determine how this molecular activity may be enhanced through the jet-to-jet coupling effect. This type of action can be used to energize some types of plasma discharge with reactive species normally not generated in them.

For these experiments, argon gas was fed into the central tube, while the outer tubes continued to use helium. This configuration would allow for the relatively strong argon discharge to be enhanced by charged particle donation from helium. Figure 4.9 shows the results of an extensive series of tests involving a combination Ar and He plasma discharge. The center tube was fed argon starting with 400sccm and increasing by 200sccm per iteration of tests. Some results have been omitted due to negligible changes between tests. For each argon flow rate, the maximum optical intensity for both the intense and well-collimated modes was determined using helium gas flows in the outer tubes. For the final argon flow rate of 1800sccm, the well-collimated behavior was not possible to obtain: the central plasma plume was not stable enough to contact the ITO glass plane in the presence of high velocity He flows.

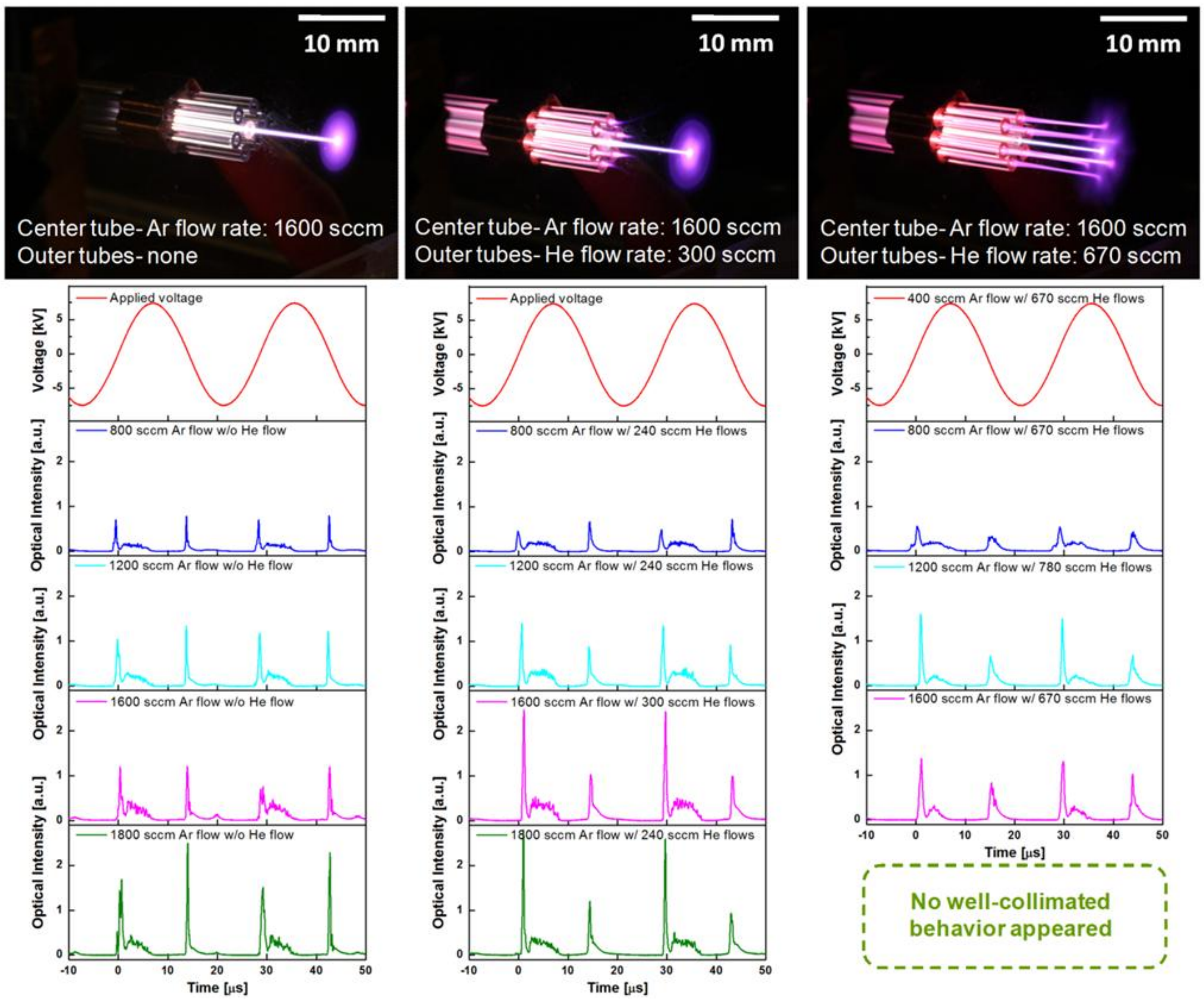

Figure 4.9: Optical intensity of Ar-He plasma driven at 15kV p-p and 35kHz. The results depict single jet (left), intense (middle), and well-collimated (right) modes of operation.

The Ar-He plasma exhibits a similar trend to the all helium plasma. Maximum possible optical intensity increases with center tube flow rate until a certain point, after which increased turbulence only detracts from the point-to-plane discharge stability. The optimal point in this data is observed to be an argon flow of 1600sccm coupled with a helium flow of 1800sccm. For this instance, the average optical intensity did not improve as radically as the all-helium discharge, but the peak of the short-lived argon excitation did increase significantly. In most cases, addition of helium plasma to the argon plume caused an increase in low-intensity emissions observable at the minima and maxima of the excitation voltage.

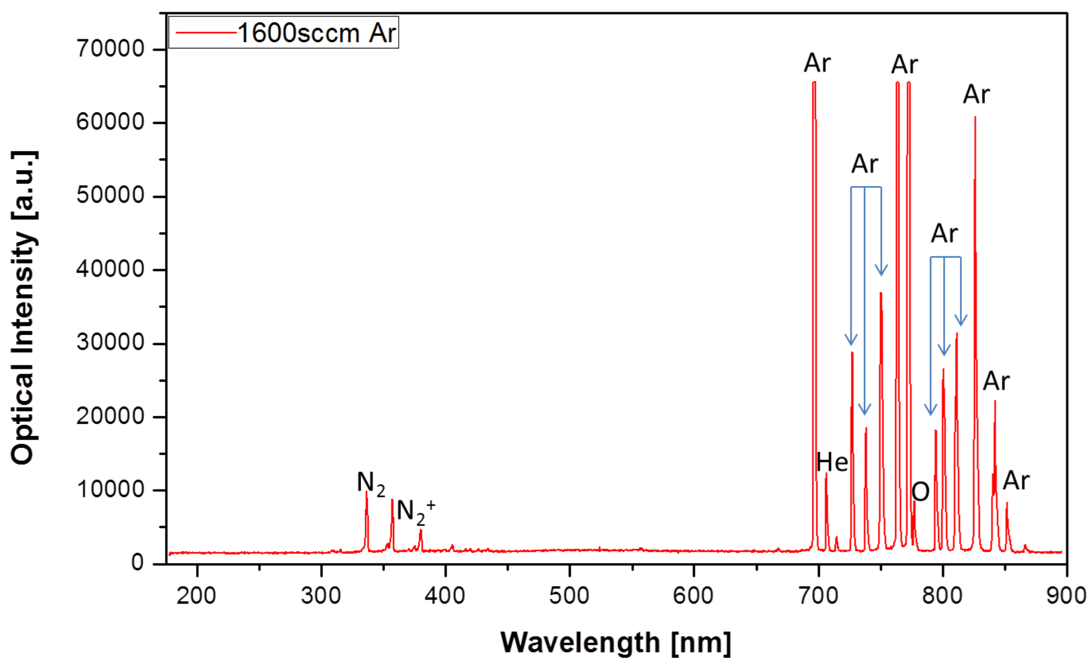

Figure 4.10: Optical emission spectrum of argon plasma. Each major peak has been labeled with its corresponding particle.

Before analyzing the contrasts between the helium and argon discharge behavior, the optical emission spectrum of argon plasma was studied. Unlike helium plasma, a large amount of the optical emission comes from excited argon atoms. Comparatively, the nitrogen and other atmospheric gases contribute little to the overall optical intensity of the plasma. Figure 4.10 shows the OES data for 1600sccm of argon flowing alone. The three tallest argon peaks in this data set exceed the sensitivity of the spectrometer used in these experiments. This was remedied with the addition of a neutral density filter in between the plasma plume and the spectrometer's photo sensor. The filtered OES results for the most intense Ar-He plasma in these experiments are shown in Figure 4.11.

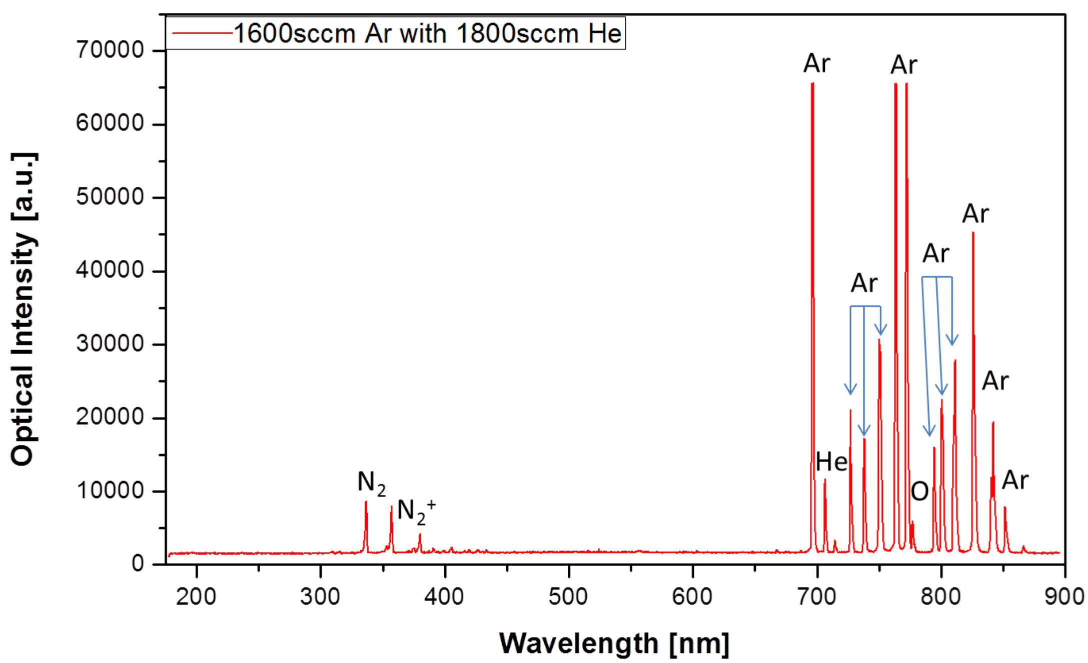

Figure 4.11: Optical emission spectrum for intense mode Ar-He plasma. This data was recorded through an ND4 filter. As a result, the scale on this graph does not accurately represent the intensity of these peaks relative to other figures.

Interestingly, even though the intense plasma discharge was optically filtered, the three argon peaks at 696nm, 764nm, and 773nm [10] remain too large to be completely measured by the spectrometer. However, relative to the other visible peaks in the spectrum, it is clear that ionized argon is the most significant source of emissions in the multiple gas plasma. Only one helium peak remains distinguishable amongst the argon peaks at 706.5nm, and it is comparatively small. Similarly, although the molecular nitrogen spectra remain visible in the results, they contribute very little to the total radiation. Whereas these reactive species were a significant portion of the helium plasma's optical emission spectrum, they do not appear to constitute much of the energy in either argon plasma or Ar-He plasma.

The most immediately noticeable difference between this set of data and the all-helium plasma intensity is the emission pattern of argon compared to that of helium. Operated under the same conditions, helium plasma demonstrates a gradual increase in optical intensity beginning with the rising and falling slopes of the excitation. On the other hand, argon plasma exhibits a sharp increase and rapid decay in energy during either slope of the excitation. Argon plasma has been observed to have greater energy transfer efficiency than helium plasma [8]. It is considered possible that the source of the large difference in recorded optical intensity for the similarly-driven plasmas is the photo sensor's greatly increased sensitivity to the larger wavelengths of light generated via argon excitation (seen in Figure 3.2). This combination was not pursued for further experimentation. The variance in behavior of these gases is likely due to argon's greater mass and lower stability at the driving conditions used. Additionally, the larger argon ions are less mobile than helium ions and believed to be suboptimal for use as ancillary feeds for helium plasma. Although it was proven that it is possible to couple dissimilar gases using this system, the extreme difference in discharge characteristics between helium and argon will most likely continue to result in poor overall improvement of the stronger plasma's properties.

4.4 Plasma Etching

The spectroscopy results of the intense helium plasma suggest that it possesses greater electron energy than either a single jet or a well-collimated array. For the purpose of etching, helium plasma is a rare consideration. Compared to heavier gases, helium plasma does not produce strong ion bombardment, and is difficult to safely energize to a point of efficacy in an etching process. Figure 4.12 shows the process of etching for an intense plume of helium plasma applied to an etching process.

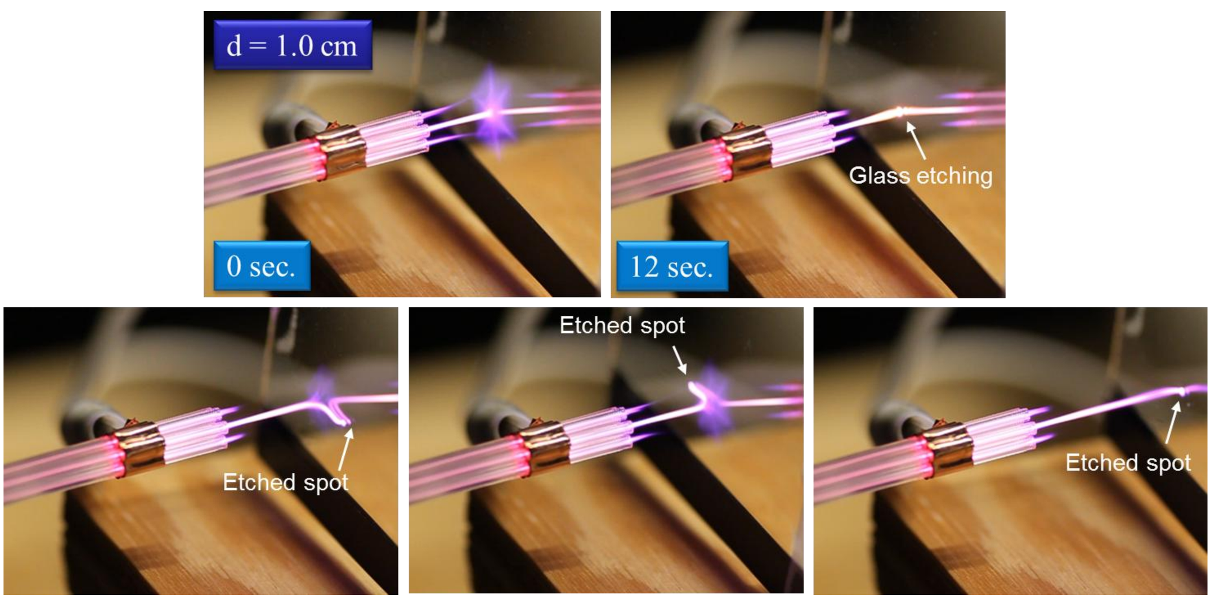

Figure 4.12: Etching process using intense helium plasma.

The etching process begins with an increase in the peak voltage of the excitation by about 5kV. Once this has been done, the plasma is usually energetic enough for ion bombardment of the surface to begin removing atoms. For helium plasma, such an increase in the excitation voltage would normally be insufficient to induce the etching behavior. For the intense plasma generated by the seven tube array, it was believed that etching would be within the realm of possibility without inordinate increase in power consumption.

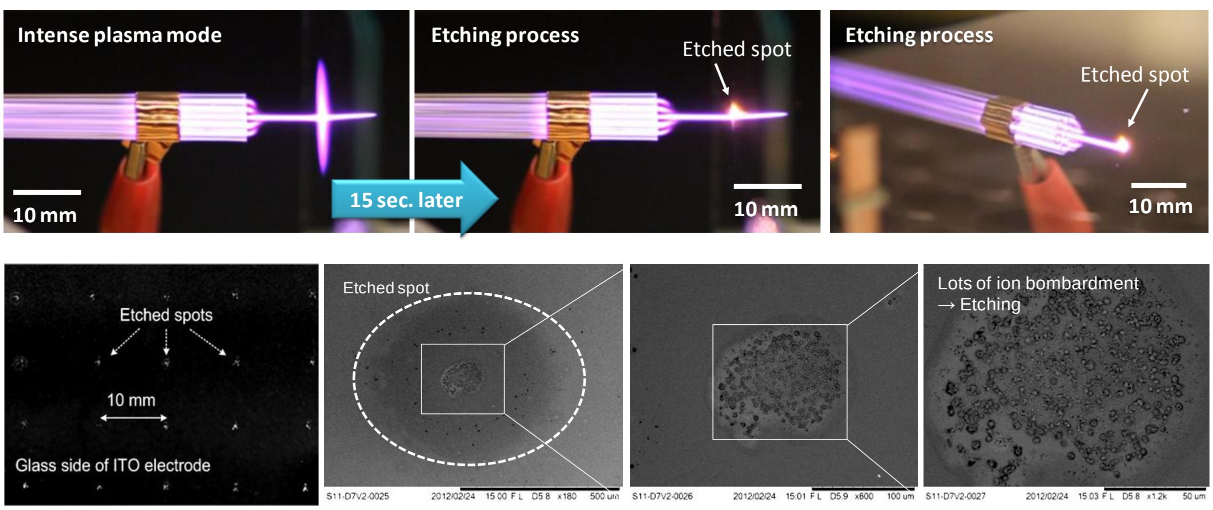

Figure 4.13: Plasma etching of ITO glass and SEM images of etched spots.

In order to test the energy of the plasma, glass etching was tested to see how quickly, if at all, the helium plume could etch into the glass side of an ITO plate. Even though the plasma system consumes less than 30W of power, the intense plume was successful in quickly etching the ITO glass. After 15 seconds of contact with the same spot, the plasma began to etch into the glass plate. When this process begins, the typical circular reflection on the glass surface disappears, as can be seen in the top middle image of Figure 4.13. After relocating the intense plasma discharge to a clean section of glass, the etching process quickly began anew. In short order, the surface of the glass plate was covered by a grid of etched spots.

SEM imaging confirms the plasma etching the ITO glass. The bottom row of images in Figure 4.10 shows magnified images of the glass surface at an etched location. Normally, in helium plasma, the ion bombardment of a surface does not impart enough energy to etch the surface. This is due to the low energy contained in the small helium ions, and their inability to cause collision cascades with enough energy to surpass a material's surface binding energy. The rightmost picture in the row shows evidence of high energy ion bombardment. Such results are generally not possible with helium plasma produced by a single jet system. These results confirm the highly energetic state of the intense-mode helium plasma.

CHAPTER FIVE

CONCLUSIONS

This work details the efforts to create intense and highly energetic atmospheric pressure plasma using an APPJ array. The effects of varying gas velocity and flow rate on plasma intensity were studied in order to produce the most intense He and He-Ar plasma possible for the array. First, the tests demonstrated that for fixed device geometry, driving conditions, and distance from the ground plane, there exists an optimum combination of gas flow rates which will produce highly energetic helium plasma. The plasma emissions were analyzed by a spectrometer in order to determine the most common charged particles in each type of plasma. The array was then tested with a combination of argon and helium to observe how the jet-to-jet coupling behavior was affected by dissimilar gases. Much like with the pure helium plasma, the Ar-He plasma exhibited an intense plasma mode with the appropriate gas settings. However, unlike the helium plasma, the mixture's maximum optical intensity was not a significant improvement over the emissions of a single jet. This is due to the innately stronger discharge of argon plasma. A mixture of similarly-behaved gases would likely provide more desirable results. The capacity of the APPJ array to etch glass was tested using the most intense Ar-He combination observed. Rapid etching of ITO glass proved that the hybrid plasma discharge proved to be just as intense and energetic as the prior test results suggested. These results certainly warrant further research into concentrating atmospheric pressure plasma into more energetic and useful forms.

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APPENDIX A: SPECTRAL DATA

Figure A.1 and Figure A.2 are a complete set of the spectral data obtained for all-helium plasma.

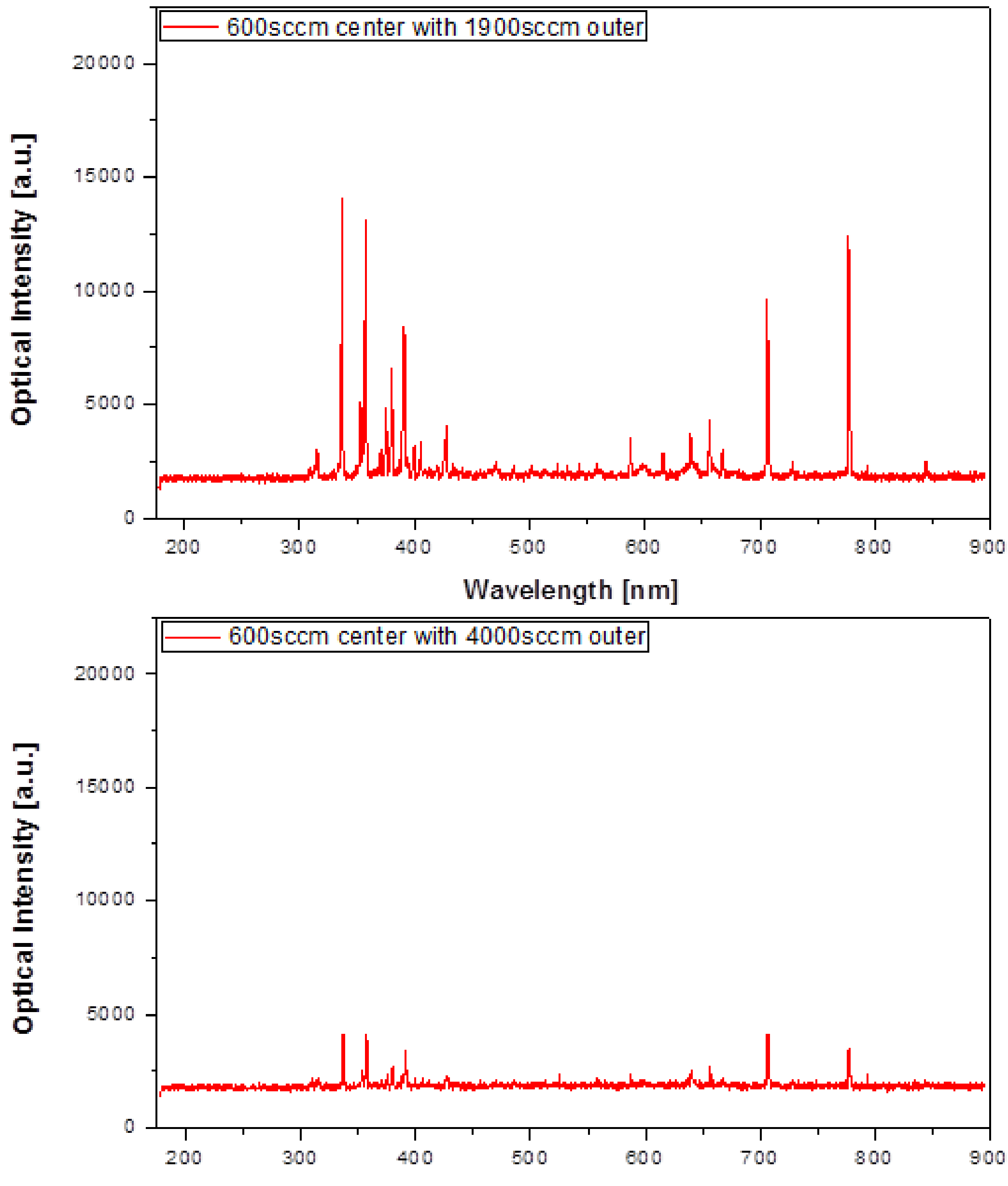

Figure A.1: Spectra for helium plasma at max intensity (top) and well-collimated mode (bottom) with a central flow of 600sccm.

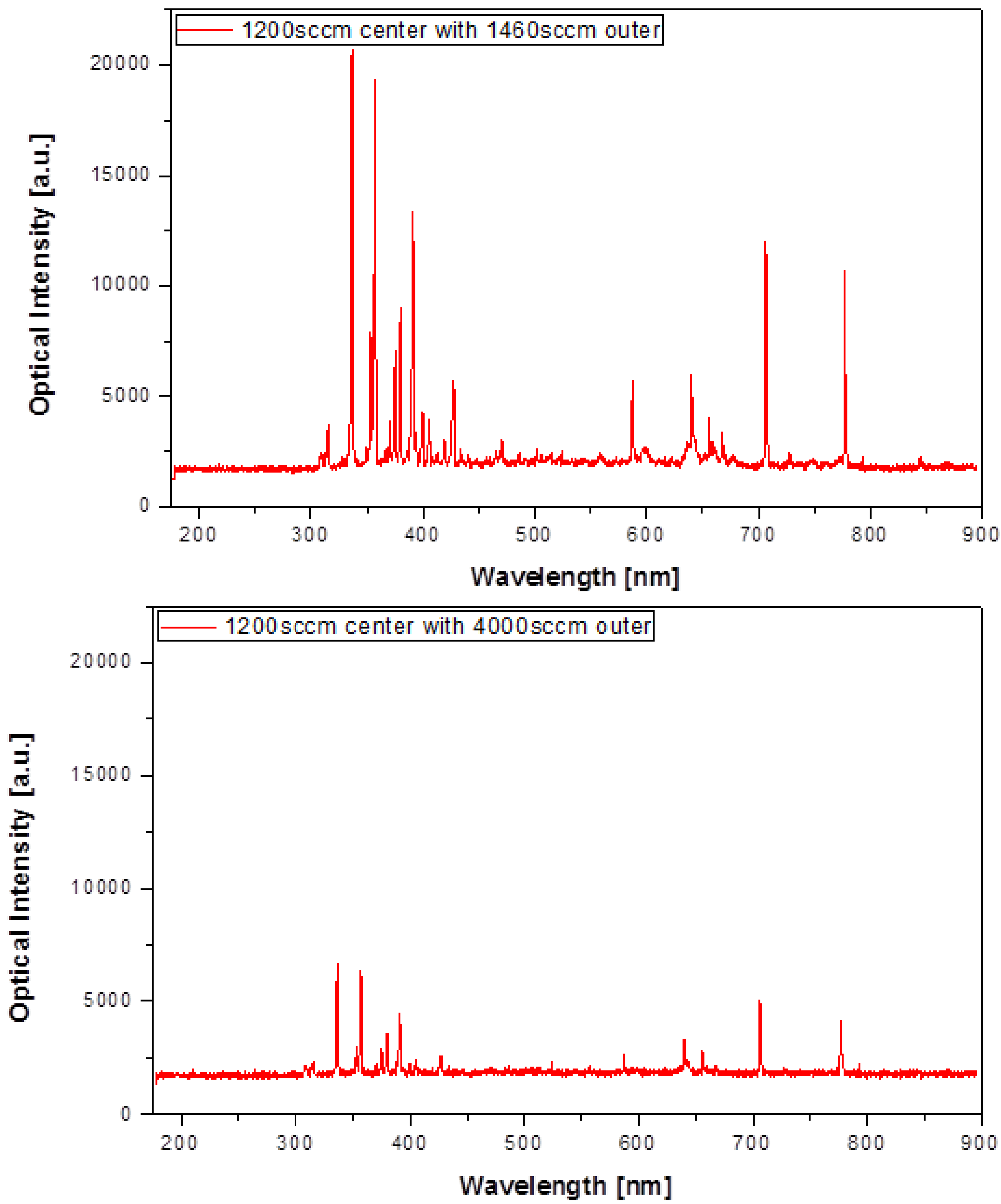

Figure A.2: Spectra for helium plasma at max intensity (top) and well-collimated mode (bottom) with a central flow of 1800sccm.

Figures A.3 through A.5 are the complete set of the spectral data obtained for the argon and argon-helium plasma.

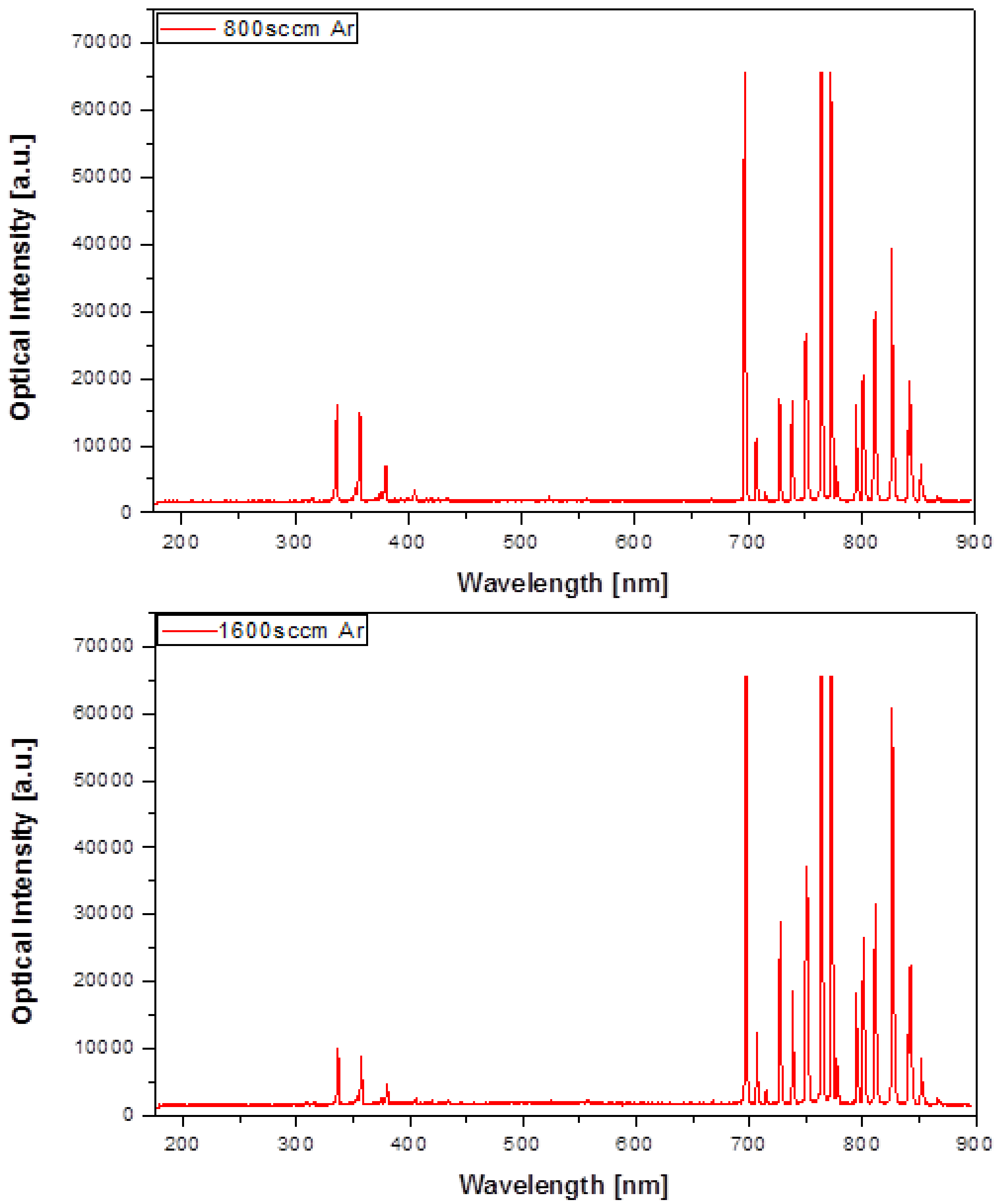

Figure A.3: Spectra for single jet argon plasma at 800sccm (top) and 1600sccm (bottom).

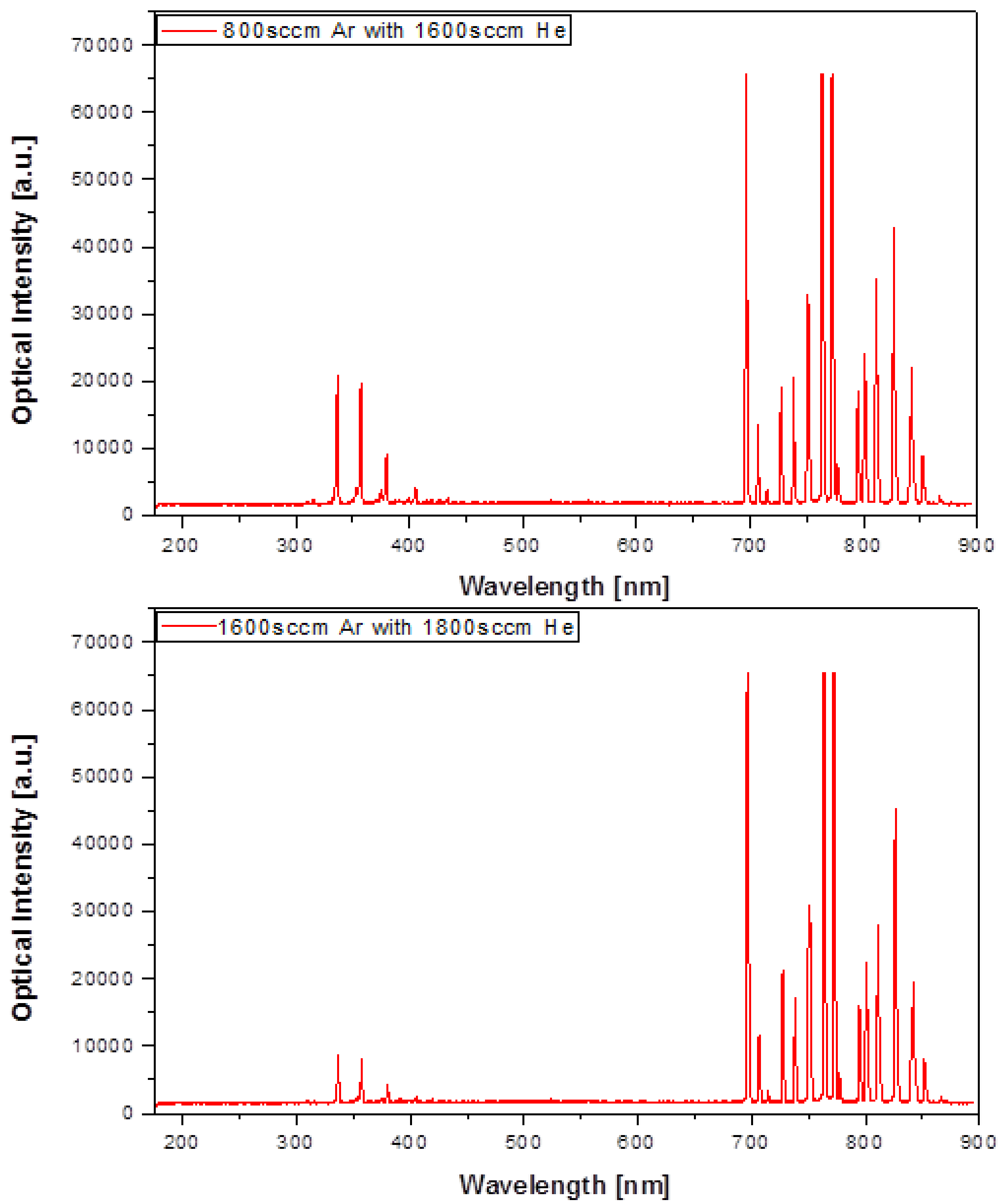

Figure A.4: Spectra for argon plasma at 800sccm max intensity (top) and 1600sccm max intensity (bottom) with outer helium flows of 1600sccm and 1800sccm respectively.

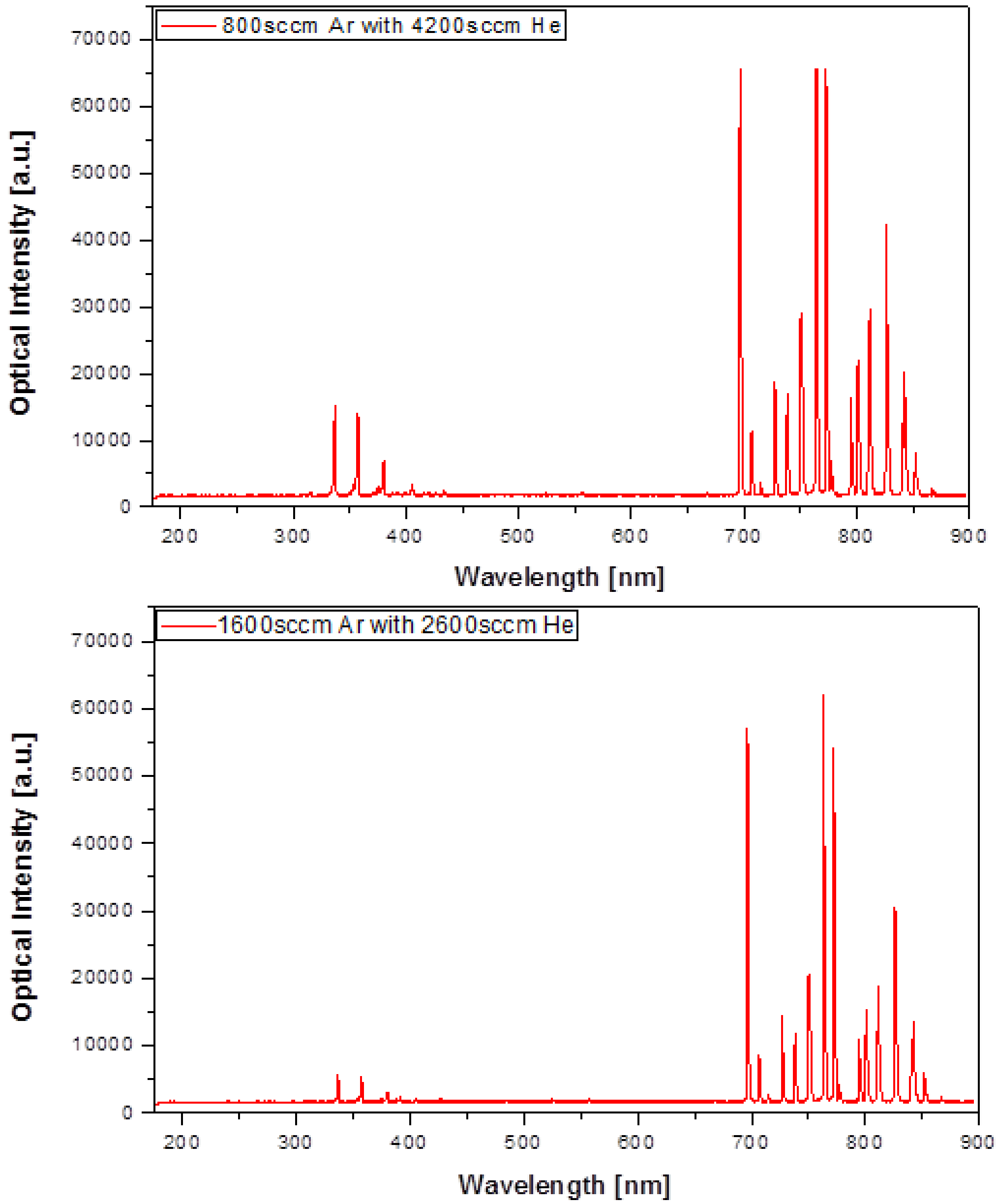

Figure A.5: Spectra for argon plasma at 800sccm well-collimated (top) and 1600sccm well-collimated (bottom) with outer helium flows of 4200sccm and 2600sccm respectively.