Abstract: Atmospheric pressure plasma jets (APPJ) are being investigated as a source of radicals and ions for surface treatment in biomedical applications. The parameter space for optimizing the reactive fluxes is large, and includes gas mixtures, voltage, gas shrouds and device-to-surface distance. Here we computationally investigate the dependence of the performance of helium APPJ on the discharge tube diameter and ground placement.

Keywords: plasma jets, ionization waves, atmospheric-pressure plasmas

1. Introduction
Recent advances in low temperature atmospheric pressure plasmas (APPJs) have broadened potential applications. Plasma jets interacting with biological liquids and tissues may be effective for cancer treatment, improved wound healing, and sterilization of heat sensitive items [1, 2]. A plasma jet typically consists of a dielectric tube with a rare gas flow seeded with a reactive gas (such as $O_2$), one or more ring electrodes outside the tube, and possibly a central pin electrode. The interaction of APPJ with liquids and tissues is through the flux of photons, ions, electrons, reactive neutral radical species, and electric fields delivered to the surface. Control of the flux of reactive oxygen and nitrogen species (RONS) to the surface is critical to optimizing their influence on various biological pathways [3]. The RONS arise from either the reactive gas in the flow or interaction of the plasma plume with the ambient air. The parameter space for optimizing and controlling these reactive fluxes is large, and includes gas mixtures, voltage, gas shrouds, device-to-surface distance and geometry. Here we computationally investigate the influences of diameter of the discharge tube and placement of the ground plane on the reactive fluxes to liquid and solid surfaces in an APPJ sustained in $He/O_2$ mixtures flowing into ambient air.

2. Description of the Model
The model used in this investigation nonPDPSIM, is a 2-dimensional plasma hydrodynamic simulation [4]. The model addresses Poisson’s equation, charged and neutral particle transport and photon transport. A modified version of the Navier-Stokes equations are solved for the bulk gas flow. The electron transport coefficients used in the continuity and electron energy equation are obtained from electron energy distributions produced by stationary solutions of Boltzmann’s equation. The calculation is performed on an unstructured mesh utilizing many refinement zones having a higher density of nodes in the plasma region. The reaction mechanism used in this analysis addresses $He/O_2$ mixtures flowing through a capillary tube into an ambient of humid air, and includes 51 species and 660 reactions.

3. Plasma Jets into Humid Air
This investigation addresses the consequences of geometry on the performance of APPJs sustained in $He/O_2$ mixtures. The base case geometry used in this investigation is shown in Fig. 1. The device consists of a cylindrical tube having an inside diameter of 1 mm and wall thickness of 500 µm. One ring electrode, 1.5 mm in height is powered. In some cases, a grounded ring electrode is also used, separated from the powered electrode by 1.5 mm. Even if research grade helium is used in the flow, there are still some impurities that are either in the as delivered gas, or introduced in the experiment. To capture some of those effects, the He flowed through the tube at 4 slm has 10 ppm $O_2$ impurity. Humid air ($N_2$/$O_2$/$H_2$O = 79.5/20/0.5) is flowed outside the tube at 4 slm. The discharge is initiated with a charge neutral spot of plasma 1 mm in diameter having a density of $1 \times 10^{12}$ cm$^{-3}$. The timescale of the gas flow is much longer than that of the discharge. In order to establish the flow profile, the model is executed with only neutral gas flow for 5 ms before the applied voltage is turned on. The pre-pulse flow field of He is shown in Fig. 1.

A negative 10 kV DC bias with a pulse length of 100 ns and a 5 ns rise time was applied to the powered electrode. The resulting electron temperature and density are shown in Fig. 2. The initial cloud of seed electrons is accelerated upward in the form of a space-charge enhanced ionization wave which propagates toward the grounded pump. The details of this propagation are specific to a negative applied bias and the placement of the ground plane. As the wave propagates, a negative surface charge builds up on the inner surface of the dielectric tube. The regions of high electron density act as a conductive, lossy dielectric that shorts out the applied voltage and extends the applied potential towards the leading edge of the ionization wave.
This extended potential, combined with space charge separation, produce electric field enhancement which is most significant on the upstream side of the plasma.

Fig. 1. (Left) Schematic of 1 mm inner diameter plasma jet. (Right) The density of helium shows the steady state flow.

At the top of the plume at 80 ns there is a conical shape to the electron temperature profile. This shape is due to the gradient of the gas composition. As the He plume extends into the ambient air, the density of air diffusing into the plume varies by several orders of magnitude. For example, 1 mm above the end of the tube, the air concentration on axis is $2.9 \times 10^{16}$ cm$^{-3}$ at the center and $2.4 \times 10^{18}$ cm$^{-3}$ at a radius of 500 µm. The air, as a molecular gas with many possible inelastic collisions, rapidly lowers the electron temperature. It is the reduction in $T_e$ by the in-diffusing air that produces the guided-streamer effect. This shape occurs only for low Reynolds number flow, as turbulence would result in dynamic air concentrations outside the tube.

The electron density just downstream of the tube continues to rise after the bias is turned off at 100 ns. No new ionization occurs after 100 ns since the electric field is too low to sustain ionization and the electrons rapidly thermalize. Rather the existing electrons are focused toward the center of the channel by residual electric fields remaining from space charge. As the ionization wave propagates, there is a net negative charge at the front edge of the streamer and a net positive charge in the bulk plasma. When the applied field is turned off, this charge separation drives the more mobile electrons back toward...
the bulk plasma, raising the electron density and the electron temperature. This occurs because the time which the voltage is turned off is shorter than the dielectric relaxation time of the plasma.

Some of the key precursors to RONS formation are the short-lived reactive neutrals formed in the plasma. The densities of He* (sum of all He excited states), O atoms, N atoms, and N$_2^*$ (sum of N$_2$(A,B)) 100 ns after the discharge begins are shown in Fig. 3. N and N$_2^*$ are formed at the interface between the ionization wave propagating in the He dominated channel and the in-diffusing air. Even with a small impurity of O$_2$, the O atom production is dominated by the higher plasma density and temperature inside the tube.

Based on these precursors, OH, and NO then form at this interface during and after the discharge. The density of OH peaks at $4 \times 10^9$ cm$^{-3}$ at 500 ns after the discharge due primarily from production by $e + H_2O^+ \rightarrow H + OH$ and $O^* + H_2O \rightarrow OH + OH$. The NO densities peak at $2 \times 10^{12}$ cm$^{-3}$ at 1.9 $\mu$s after the discharge due to production by $O + N_2^* \rightarrow NO + N$.

The flow of the atomic oxygen interacting with the ambient gas contributes to RONS formation. Atomic oxygen densities from the impurity oxygen in the gas flow peak at $6 \times 10^{12}$ cm$^{-3}$ inside the tube at the end of the discharge pulse. After the discharge extinguishes, the flow carries the atomic oxygen out of the tube to interact with the ambient O$_2$, rapidly forming O$_3$ which reaches a density of $3 \times 10^{11}$ cm$^{-3}$ at 25 $\mu$s after the discharge extinguishes.

Decreasing the diameter of the tube without changing the flow rate decreases the residence time of the gas in the jet, but will not significantly change the total energy deposition. Changing the diameter of the tube should not have a dramatic effect on the total inventory of reactive species produced (the electrons, ions, and relevant reactive neutrals). However, more rapid mixing with the ambient air may alter the resulting RONS. Grounding the downstream ring provides a more local ground than the pump, which will affect the propagation of the ionization wave out of the tube. This propagation is the primary mechanism of OH and NO production in the interface between the plume and the in-diffusing air. The discharge produced inventory of these species will be more sensitive to the ground placement than the O$_3$ inventory, whose precursor, O atoms, are produced primarily in the tube.

4. Concluding Remarks
An atmospheric pressure plasma jet with a single powered ring electrode has been computationally investigated. Even at impurity levels as low as 10 ppm, oxygen in the flow gas can have a significant effect on the O$_3$ production of the jet. As the discharge exits the jet, the profile of the electron temperature is determined by the in-diffusion of air into the plasma plume. OH and NO are primarily produced at the interface of the ionization wave and the in-diffusing air. O$_3$ is also produced at this interface when the atomic oxygen produced during the discharge flows out into the ambient. The effect of tube diameter on discharge dynamics will be discussed, and a comparison will be made between discharges with a grounded pump and grounded downstream ring electrode.

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6. References