Investigation of the Production of O$_2$(a$^1\Delta$) in Rectangular and Multi-Tube Radio-Frequency Discharges


University of Illinois at Urbana-Champaign, Urbana, IL 61801

CU Aerospace, Champaign, IL 61820

Abstract

Experiments and modeling have led to continued enhancements in the Electric Oxygen-Iodine Laser (ElectricOIL) system. This continuous wave (cw) laser operating on the 1315 nm transition of atomic iodine is pumped by the production of O$_2$(a) in a radio-frequency (RF) discharge in an O$_2$/He/NO gas mixture. New discharge geometries have led to improvements in O$_2$(a) production and efficiency. Studies of electrode gap continue to improve O$_2$(a) production at high pressures. Additionally, measurements of species exiting the discharge and the determination of discharge parameters such as E/N continue to expand the understanding of this system. Some of these improvements have already been applied to the laser system, and other advances will be utilized to continue scaling the system to higher laser powers. Over 1 kW of power stored in O$_2$(a) has been demonstrated in both the rectangular cross-section and multi-circular tube discharges.

I. Introduction

The electrically driven oxygen-iodine laser (ElectricOIL) that was first demonstrated by Carroll et al.\textsuperscript{1,2} operates on the electronic transition of the iodine atom at 1315 nm, I($^3P_{1/2}$) $\rightarrow$ I($^3P_{3/2}$) [denoted hereafter as I$^\ast$ and I respectively]. The lasing state I$^\ast$ is produced by near resonant energy transfer with the singlet oxygen metastable O$_2$(a$^1\Delta$) [denoted hereafter as O$_2$(a)]. Since the first reporting of a viable electric discharge-driven oxygen-iodine laser system (also often referred to as EOIL or DOIL in the literature), there have been a number of other successful demonstrations of gain\textsuperscript{3,4,5} and laser power\textsuperscript{4,5,6} as well as a recent demonstration of gain and lasing from an air-helium discharge.\textsuperscript{7} Computational modeling of the discharge and post-discharge kinetics\textsuperscript{8,9,10} has been an invaluable tool in ElectricOIL development, allowing analysis of the production of various discharge species [O$_2$(a$^1\Delta$), O$_2$(b$^1\Sigma$), O atoms, and O$_3$] and determination of the influence of NO$_3$ species on system kinetics. Ionin et al.\textsuperscript{11} and Heaven\textsuperscript{12} provide comprehensive topical reviews of discharge production of O$_2$(a) and various ElectricOIL studies. As of approximately one year ago\textsuperscript{13}, the highest reported gain in an ElectricOIL device was 0.2 \% cm$^{-1}$, with an output power of 28.1 W, and currently the gain and laser power stand at 0.26 \% cm$^{-1}$ and 102 W, respectively.\textsuperscript{14}

O$_2$(a) excitation in an electric discharge is a complicated process. Optimization requires a discharge with E/N (electric field-to-gas density ratio) favoring production of the desired O$_2$(a) state which has an electron-impact threshold at 0.977 eV above the ground state. The discharge E/N is characteristic of the electron energy distribution function which describes the state of the electron gas within the plasma. Regardless of how well this distribution is controlled to optimize for O$_2$(a) production, the distributed energy of the electron gas results in a variety of processes taking place within the discharge, and the gas flow exiting the discharge region is not simply a flow consisting primarily of O$_2$(a) and O$_2$(X) (as in classic COIL singlet-oxygen generation). Instead, the discharge energy is deposited into a variety of states, which include (i) vibrational and rotational energy, (ii) thermal energy (gas temperature), (iii) energy stored in electronic energy states of molecules and atoms, and (iv) dissociated molecules. This complicated situation must be dealt with accordingly. The discharge cannot be treated as a “black-box” which produces significant O$_2$(a) for sustaining a population inversion in atomic iodine. It is also necessary to

---

1 Res. Assistant, Univ. of Illinois at Urbana-Champaign, 104 S. Wright, Urbana, IL 61801, Student Member AIAA.
2 Senior Engineer, CU Aerospace, 2100 S. Oak St. – Ste 206, Champaign, IL 61820, Member AIAA.
3 Senior Physicist, CU Aerospace, 2100 S. Oak St. – Ste 206, Champaign, IL 61820, Senior Member AIAA.
4 Vice President, CU Aerospace, 2100 S. Oak St. – Ste 206, Champaign, IL 61820, Associate Fellow AIAA.
5 Senior Scientist, CU Aerospace, 2100 S. Oak St. – Ste 206, Champaign, IL 61820.
6 Prof. Emeritus, Univ. of Illinois at Urbana-Champaign, 104 S. Wright, Urbana, IL 61801, Fellow AIAA.
understand how energy is transferred into the other processes in the discharge and how the “byproducts” of these processes influence the overall system kinetics, primarily the maintenance of \( \text{O}_2(a) \) levels in the system, and the transfer of the energy stored in \( \text{O}_2(a) \) into the \( ^1 \) laser state.

With the transverse RF discharge chosen as the most suitable discharge configuration for ElectricOIL development by the Univ. of Illinois and CU Aerospace team, a detailed configuration study of the influence of various discharge parameters on \( \text{O}_2(a) \) production was conducted. The parameters that proved important in optimizing \( \text{O}_2(a) \) production were (i) operating pressure, (ii) transverse gap, (iii) discharge flow length, and (iv) excitation frequency, consistent with the findings of Braginsky et al.\(^{15} \) The key result of the work was that in order to achieve efficient production of \( \text{O}_2(a) \), a uniform, normal-mode glow discharge must be maintained. In the normal-mode glow discharge, low current density is maintained, and as more power is applied to the discharge, the volume of plasma in the tube between the electrode plates increases proportionally, maintaining a similar current density for increased power deposited while voltage remains about constant. If the plasma fills the volume between the electrode plates and is confined, the discharge is then forced into an abnormal mode in which the plasma voltage and current density rise in unison. This simple transition from a normal to an abnormal mode can severely decrease the \( \text{O}_2(a) \) production efficiency. However, the onset of this transition can be delayed by careful selection of the transverse discharge parameters (i) – (iv). A variety of diagnostics were used to study the dynamics of this transition including various excited state measurements, airglow (O-NO recombination) and actinometry measurements, plasma glow distribution measurements, and plasma density and collision frequency measurements using microwave interferometry.

II. Experimental Apparatus

All the results discussed in this paper were produced using a capacitively coupled transverse radio-frequency (RF) discharge operating at 13.56 MHz. Two different types of discharge tube cross-sections were utilized, and Fig. 1 shows the cross-sections that were used during the high flow rate experiments. All the discharge tubes are made of quartz, and the rectangular tubes were custom fabricated. One rectangular tube was 24 inches long, and another was 48 inches long. Unfortunately, after repeated use in the vacuum system both edge-welded tubes failed irreparably. The circular cross-section tubes are standard parts with an outer dimension of 19 mm (¾ in.) and inner dimension of 16 mm. This research used many lengths of those circular tubes up to 48 inches. Both designs shown in Fig. 1 have an internal cross-sectional area of about 1200 mm\(^2 \). Obviously, the surface area per unit length is greater in the six tube design. During lower total flow rate experiments, a single circular tube was used with parallel plate electrodes. Other single tube tests were conducted with a 12 mm (½ inch) OD tube with an ID of 10 mm. The length of the electrodes varied by experiment. All of the tubes in this work are quartz except for the clearly marked cases where the tube was made from alumina. For the single ¾ and ½ inch OD tube experiments, the gas flowed out of the discharge tube and into a 1 inch diameter, 18 inch long tube before entering a diagnostic block with purged windows. Measurements of \( \text{O}_2(a), \text{O}_2(b^1 \Sigma) \) [denoted hereafter as \( \text{O}_2(b) \)], and \( \text{NO}_2^* \) emission along with absorption of ozone were made in this setup. The 18 inch tube separating the discharge from the diagnostics was required to minimize RF interference with the ozone absorption diagnostic. In the high flow rate cases where the discharge was either one of the configurations shown in Fig. 1, a 2 inch diameter, 18 inch long tube separated the discharges and the purged diagnostic block where the same emission measurements were made. In this case, the extra length was required to allow the flow to diffuse and uniformly fill the diagnostic volume after exiting discharges with quite different cross-sections.

![Figure 1. Cross-sectional illustration of the quartz rectangular tube and multi-circular tube discharge configurations with electrodes. The high voltage (HV) and ground electrodes are indicated. The flow direction is into the paper, and the sketch is not to scale.](image-url)
An ENI OEM-25A or ENI OEM-50 provided the power for the experiments at 13.56 MHz, and the incident and reflected powers to the RF matching network were measured by a Bird Thruline model 43 wattmeter (“RF Power” in the figures is the difference between the incident and reflected powers). Matching the power to the discharge was achieved using a traditional PI-matching network. The same variable inductance matching network was utilized for both configurations shown in Fig. 1. Micro-Motion CMF and Omega FMA mass flow meters were used to measure the flow rates of the gases. Pressures in the flow tubes were measured with MKS Instruments and Leybold capacitance manometers. Princeton Instruments/Acton Optical Multi-channel Analyzer (OMA-V, 1024-element InGaAs array) with a 0.3 m monochromator and a 600 g/mm grating blazed at 1 µm was used for measurements at 1268 nm. An Apogee E47 CCD camera coupled to a Roper Scientific/Acton Research 150-mm monochromator was used to measure the emission of O$_2$(b) at 762 nm to determine flow temperature. The oxygen atom concentration was determined using broadband emission of NO$_2^*$, measured using a Hamamatsu R955 photomultiplier with a narrowband 580 nm filter and a 50 mm focal length collection lens; the oxygen atom concentration was determined from NO$_2^*$ using the method described by Piper.$^{16}$ These optical diagnostics were fiber coupled using either Oriel model #77538 glass fiber bundles or ThorLabs 600 µm x 5 m multimode fibers. The ThorLabs fiber used for the O$_2$(a) emission measurement was connected to a ThorLabs F810SMA-1310 collimator that attaches to the diagnostic block.

Throughout this work, O$_2$(a) yields from a variety of RF discharges are compared. The proportions of oxygen to helium are carefully controlled and, with the exception of one data series, are always 1:3.33, O$_2$:He. The addition of NO to the discharge eliminates oxygen atoms through a recycling process and boosts the production of O$_2$(a).$^{17}$ These effects are shown in Fig. 2. This plot was created using the rectangular cross-section quartz tube with 45:150 mmol/s, O$_2$:He at 40 Torr and 4000 W of RF power. An NO flow rate ranging from 0.5% to 1.5% of the oxygen flow rate produced the same effect on the O$_2$(a) while causing substantial impact on the oxygen atoms. Energy is released as the oxygen atoms recombine, and this is illustrated by the increasing temperature as more NO is added to remove more atoms. Subsequent comparisons of O$_2$(a) are made with inconsistent NO flow rates, but they are within the range where the O$_2$(a) yield is not affected. In the few cases where oxygen atom yields are compared, the NO flow rate was closely controlled. A modest, 9% increase in O$_2$(a) yield occurs in this configuration when NO is added to the flow. Previous ElecticOIL studies illustrated a substantial increase in O$_2$(a) yield of almost 30% when NO was added to the system using a longitudinal discharge at 13 Torr.$^{17}$ As discharge operating pressures have increased with the use of transverse, small electrode gap discharges, the need for NO to produce high O$_2$(a) yields has decreased while NO remains critical for oxygen atom control.

III. Experimental Results

A. Ozone Measurements

Understanding the role of oxygen species created in the discharge that are not present in classic COIL has been critical to the success of ElecticOIL. Oxygen atoms proved to be critical players in the chemistry due to their positive role dissociating iodine and their negative role quenching the upper lasing state.$^{18}$ Ozone is also created in the discharge, and studies at lower pressures (~15 Torr) and flow rates with large electrode gap discharges showed
that the concentration of ozone was small compared to the other species.\textsuperscript{10} Discharges operating at higher pressures with smaller electrodes gap have primarily been studied and optimized for their $O_2(a)$ production. Ozone is produced through several three-body reactions (shown as Eqns. 1-4)\textsuperscript{19}, so its production could increase drastically at higher pressure operation.

\begin{align*}
O + O_2 + He & \rightarrow O_3 + He \quad (1) \\
O + 2O_2 & \rightarrow O_3 + O_2 \quad (2) \\
O + O_2 + O_2(a) & \rightarrow O_3 + O_2(a) \quad (3) \\
2O + O_2 & \rightarrow O_3 + O \quad (4)
\end{align*}

These ozone production reactions are assumed to have rates of $5.9 \times 10^{-34} \text{ cm}^6/\text{molecule}^2\text{-s}$ at room temperature. The negative effects of ozone on the ElectricOIL system are shown as Eqns. 5 and 6.\textsuperscript{19}

\begin{align*}
O_2(a) + O_3 & \rightarrow 2O_2 + O \quad k = 4.02 \times 10^{-15} \quad (5) \\
I + O_3 & \rightarrow IO + O_2 \quad k = 1.03 \times 10^{-12} \quad (6)
\end{align*}

The units of these reactions are $\text{cm}^3/\text{molecule}\text{-s}$, and they are also evaluated at room temperature. Clearly, deactivation of $O_2(a)$ and removal of iodine atoms are serious problems for an ElectricOIL system, so the concentration of ozone needed to be determined.

Measurements of ozone concentration are performed using an absorption diagnostic built by Physical Sciences Inc. This tool is capable of measuring concentrations of ozone as low as $1.7 \times 10^{11} \text{ cm}^{-3}$ with a 5 cm path length using an ultraviolet light source.\textsuperscript{20} The experiment was configured to match the discharge conditions used during recent laser testing that produced high output laser powers.\textsuperscript{21} A single $\frac{3}{4}$ inch tube was used with the flow rates and RF power scaled to 1/6 of their laser test values. The diagnostic utilized a 2.5 cm path length and was located approximately 19 inches downstream from the exit of the discharge. The NO flow rate was swept for several discharge pressures to produce Fig. 3. The yield of ozone, defined as $[O_3]/[O_2]$ input, is plotted against the NO flow rate rather than the number density since the pressure changes drastically between data cases making the number density of ozone a confusing comparison. All the ozone production reactions require oxygen atoms, so for all pressures, the ozone yield is the highest in the case with zero NO flow. NO is recycled to eliminate oxygen atoms, and as the pressure increases, the oxygen atoms recombine faster. For the case without NO, the ozone yield reaches a maximum at 60 Torr before the loss of oxygen atoms at high pressure reduces the yield at 80 Torr. With the addition of NO to the flow, the density of ozone drops to about $1 \times 10^{14} \text{ cm}^{-3}$ for all pressures. More oxygen atoms remain at 20 Torr providing a higher yield at all NO flow rates while the yield at the three higher pressures falls substantially. In order to put these ozone yield and density measurements into perspective, Fig. 4 shows the number density of the most common post-discharge species as a function of pressure. These data are from the 0.13 mmol/s of NO case in Fig. 3, and all the measurements were taken at approximately the same location downstream of the discharge. This log plot shows that the concentration of ozone is substantially smaller than that of $O_2(a)$, $O_2(b)$, or oxygen atom at the measurement location. At the discharge exit, the oxygen atom density is much higher than it is further downstream, especially at high pressures.
B. Smaller Electrode Gap Experiments

As the pressure in these transverse RF discharges increases, the electrode gap for maximum $O_2(a)$ production decreases. Discharges have been created for high gas flow rates that operate well between 40 and 50 Torr with an electrode gap of approximately ¼ inch. In order to scale these types of systems to higher laser output powers, the total gas flow rates must increase. Improved pumping capabilities and diffuser design can facilitate some of that increased gas flow, but improved discharge operation at higher pressures will help reduce the dependence on those factors. Testing with ½ inch OD quartz discharge tubes has shown a substantial benefit over ¼ inch tubes at pressures above 50 Torr as Figs. 5 and 6 illustrate. In the data for both figures, the flow rates are proportional to the cross-sectional area of the tubes, so that the flow time within the discharge is the same for a given pressure. In Fig. 5, the $O_2(a)$ yield is plotted against the RF power divided by the input oxygen flow rate, and in Fig. 6, the yield is plotted versus the pressure with a constant RF power of 100 W per mmol/s of input oxygen. The top and bottom data in Fig. 5 are from the ¼ inch discharge at 40 and 80 Torr, respectively. The data between those curves is from the ½ inch tube. At 40 and 60 Torr, the smaller tube’s performance is similar and slightly lower than the $O_2(a)$ yield in the ¾ inch tube. At 80 Torr, the yield is almost twice as high in the ½ inch tube compared to the ¼ inch tube although it is still substantially worse than either of the 40 Torr cases. Figure 6 shows that at pressures above approximately 50 Torr the smaller discharge tube provides higher yield. The benefit of the small electrode gap may be even more pronounced than Fig. 6 shows. The flow time within the discharge was held constant for these tests, but the distance between the discharge exit and the measurement location was also a constant. The time for $O_2(a)$ to decay is more than double in the low flow rate case corresponding to the ½ inch tube. If measurements could be made at the discharge exit, the ½ inch tube might appear to be even better at high pressures. Higher flow rates were attempted through the small discharge tubes, but large pressure drops within the discharge tubes were observed in these cases. With 10:33 mmols, $O_2$:He, the discharge inlet pressure was about 80 Torr when the downstream pressure was only 40 Torr. These large variations in discharge pressure produced confusing results, so the oxygen flow rates through the ½ inch discharge tubes is limited to between 4 and 6 mmol/s of $O_2$ with the corresponding amount of helium where the pressure drop is only around 10 Torr.
C. Discharge Length Investigation

Simulations using the BLAZE-IY<sup>9,10</sup> discharge model suggested that, in the smaller diameter discharge tubes, the 10 inch electrodes did not provide sufficient residence time for the flow within the discharge to reach an equilibrium of O<sub>2</sub>(a) production. Experiments with a single ¾ inch discharge tube supported this conclusion<sup>11</sup>, and a long, rectangular cross-section quartz tube was fabricated to further exploit this finding for increased O<sub>2</sub>(a) production. Electrodes with lengths of 10, 20, 30, and 42 inches were placed on this tube, and the results from several gas flow rates were measured. The O<sub>2</sub>(a) yield versus the ratio of RF power to input oxygen flow rate is shown in Figs. 7 through 9. Several flow rates are shown for each electrode length, and the ratio of O<sub>2</sub>:He was held at 1:3.33 with approximately 0.3 mmol/s of NO. The discharge pressure in each of these cases was 40 Torr. From Fig. 7 to Fig. 8, the electrode length doubled, and as expected from modeling, the O<sub>2</sub>(a) yield increased by about 20%. The electrode length was doubled again to 42 inches (Fig. 9), and the yield stayed almost exactly the same as the 20 inch electrode case (Fig. 8). Data not shown here with 30 inch electrodes fall in line with the 20 and 42 inch data. With electrode lengths of 20 inches and longer, the peak O<sub>2</sub>(a) production occurs at approximately 80 W/mmols/s of O<sub>2</sub>. This result is somewhat surprising considering that with a single tube, the peak of O<sub>2</sub>(a) production with a similar electrode gap occurs at 100 W/mmols/s of O<sub>2</sub> (Fig. 5). The volume of the electrodes filled by the discharge varies with power and pressure, but all the discharges filled the entire electrode volume for the 10 inch case (Fig. 7), so the discharge residence time was already approximately double for the 30 mmol/s of O<sub>2</sub> case compared to the 60 mmol/s of O<sub>2</sub> case. Nevertheless, when the electrodes were lengthened, all the varied flow rate cases showed substantial improvement. If discharge residence time was the only contributing factor to O<sub>2</sub>(a) production, then three substantially different flow rates through the same discharge would not peak in O<sub>2</sub>(a) yield at the same electrode length. Furthermore, an experiment was performed where the discharge length was held constant at 42 inches as the input power changed. Figure 10 contains these results. This experiment was performed by adding a 1.5 inch electrode upstream of the main 42 inch electrodes and powering it with a relatively small amount of RF power. This additional “primer” discharge caused the primary discharge to fully fill the electrodes regardless of the primary input power. In this case, the primer discharge had 500 W applied to it, and all the remaining power was applied to the primary discharge. As Fig. 10 shows, the discharge did not fill the electrode volume on its own until 4200 W had been provided. With the primer discharge engaged, the discharge always filled the entire volume. For this case of 45:150:0.3 mmol/s, O<sub>2</sub>:He:NO at 40 Torr, the increased discharge volume provided no benefit. The residence time in this case is approximately 5 ms, and the previously mentioned model suggests that the O<sub>2</sub>(a) would not reach equilibrium until sometime around 40 ms at this pressure although the predicted and measured O<sub>2</sub>(a) yields are similar. The O<sub>2</sub>(a) concentration appears to be reaching equilibrium faster than the model predicts, but the reasons for that remain unknown.
Figure 7. $O_2(a)$ yield vs. RF power per input oxygen flow rate at 40 Torr with 10 inch electrodes on the rectangular cross-section tube. Approximately 0.3 mmol/s of NO in each case.

Figure 8. $O_2(a)$ yield vs. RF power per input oxygen flow rate at 40 Torr with 20 inch electrodes on the rectangular cross-section tube. Approximately 0.3 mmol/s of NO in each case.

Figure 9. $O_2(a)$ yield vs. RF power per input oxygen flow rate at 40 Torr with 42 inch electrodes on the rectangular cross-section tube. Approximately 0.3 mmol/s of NO in each case.

Figure 10. $O_2(a)$ yield and discharge length vs. RF power with 42 inch electrodes. In one case the discharge length is allowed to change, and in the other, it is held at the full electrode length. The pressure is 40 Torr, and the flow rates are 45:150:0.3 mmol/s of $O_2$:He:NO.

D. Microwave Interferometer Measurements

A microwave interferometer operating at 4.1 GHz was used to deduce electron density and collision frequency in moderate pressure plasma similar to those used in ElectricOIL experiments. The measurement was made in a transverse RF discharge with rectangular cross-section, having a 3-cm gap, a flow channel width of 6 cm, and an electrode length of 25.4 cm (in the flow direction). The microwave beam was passed through the side walls of the discharge tube, on a path perpendicular to the flow direction. HP G281A waveguide adapters (G-Band, WR-187) were used as horns to transmit and receive the microwave beam on either side of the discharge tube, as illustrated in Fig. 11. The microwave interferometer setup used is described in Fig. 12.
Figure 11. Sketch of plasma vessel and microwave interferometer “horns” setup. Microwaves propagate along the x-axis between the waveguide adapters (plane waves assumed in analysis). The microwave electric field is in the y-direction, while the magnetic field is along the z-axis. Microwaves propagate through five mediums: (0 and 4) air, (1 and 3) quartz and (2) plasma.

Figure 12. Microwave interferometer setup for measurements in a transverse discharge. The objective of the experiment was to measure the change in attenuation and phase shift of the microwave beam due to the plasma, and relate these to the plasma electron density $n_e$ and collision frequency $\nu_m$. The microwave source signal is split (by a coupler) into two signal arms, one which passes through the plasma vessel (plasma arm), and one which passes through a phase shifter and step attenuator (reference arm). These two signals are summed using a 90° coaxial hybrid. With the RF plasma off, the signals from the two arms are balanced at the hybrid, resulting in a null (zero signal due to destructive interference, signals equal with 180° phase difference) at channel 1, and a double magnitude signal (due to constructive interference, signals equal and in phase) at channel 2. Once this condition is established, the RF plasma is initiated, and the signal passing through the plasma arm is attenuated and phase shifted with respect to the reference arm signal resulting in an increase (from null) of the signal on channel 1, and a decrease in the signal on channel 2. The phase shifter in the reference arm is adjusted to minimize the signal on channel 1 (maximizing channel 2). The change in phase shifter setting required to minimize the signal gives the phase shift due to the plasma, and the signals on channel 1 and 2 are used to deduce the attenuation. Based on attenuation and phase shift measurements, the electron density and collision frequency are determined using the
method described by Bachynski and Graf, which accounts for the reflections due to the air/quartz and quartz/plasma interfaces. Results from this method in helium plasma and in 10:33 O₂:He mixture without and with NO are shown in Fig. 13. Assuming direct current conductivity, the E/N can be deduced from the results shown in Fig. 13 and gas temperature measurements. The resulting E/N ranges for the data shown were: 20-35 Td (1 Td = 1x10⁻¹⁷ V·cm²) at 5 Torr, 10:33 O₂:He; 16-22 Td at 20 Torr, 10:33 O₂:He; 7-9 Td for He only at 4 Torr.

---

**E. E/N Estimated from Current-Voltage-Characteristic (CVC)**

Previous work showed current-voltage-characteristics (CVC), the voltage across the electrodes as a function of current into the driven electrode, for a 3-cm gap transverse discharge at varied pressure. The data shown were the voltage and current as RF power varied in both the normal case (voltage and current density approximately constant) and the abnormal case (voltage and current increase in unison with power input). By accounting for the stray capacitance (in this case 23 pF) and estimating the voltage drop across the quartz dielectric and the plasma sheaths, the voltage and current information can be used to estimate the plasma E/N in the positive column (PC).

The voltage $V$ across the electrodes as a function of current density $J$ is shown in Eqn. 7.

$$V(J) = \left[ V_{\text{sheath}}^2 + V_{\text{PC}}^2 \right]^{1/2} = \left[ \left( \frac{d_1 + 2(d_q / \varepsilon_q) J}{\varepsilon_0 \omega} \right)^2 + \left( C p d_J m \right) \right]^{1/2}$$  

(7)

$V_{\text{sheath}}$ is the combined voltage across the sheath and quartz walls, $V_{\text{PC}}$ is the positive column voltage, $d_1$ is the (α-mode) sheath thickness, $d_q$ is the quartz thickness, $\omega$ is the excitation frequency, $\varepsilon_0$ is the freespace permittivity, and $\varepsilon_q$ is the quartz permittivity, $d_1 = d - d_q$ is the PC gap, and $p$ is the pressure; $C$ and $m$ are constants dependent on the gas which describe the descending character of the PC CVC. If the power form for $V_{\text{PC}}$ is used as in Eqn. 7, the function has a minimum at $J_{\text{norm}}$, shown in Eqn. 8, which corresponds to the normal current density.

$$J_{\text{norm}} = \left( \frac{\varepsilon_0 \omega C p d_J m}{d_1 + 2(d_q / \varepsilon_q)} \right)^{1/(m+1)}$$  

(8)

The modeling of the discharge CVC using Eqn. 7 is shown in Fig. 14. The E/N can be estimated by determining the positive column CVC $V_{\text{PC}}(J)$ and sheath thickness which best fit the $V(J)$ data (model in Fig. 14), and then estimating the reduced E/N as $V_{\text{PC}}(J)/(d_1+N_{\text{avg}})$, where $d_1$ is the PC gap, and $N_{\text{avg}}$ is the average gas density in the
discharge determined from pressure and the temperatures deduced from O_2(b) spectra taken within the discharge region. The E/N determined from modeling the CVC is shown in Fig. 15; the E/N values are between 20 and 25 Td, similar to the results obtained with microwave interferometer. The normal current densities (minimum of V/J in Fig. 14) for the 10, 21, and 32 Torr cases are 1.8, 5.2, and 9.2 mA-cm^2 respectively, while the sheath thicknesses are 0.19, 0.11, and 0.092 cm respectively.

Figure 14. Comparison of the measured and calculated terminal CVC for 3-cm gap transverse RF discharge, flowing 10:33 mmol/s O_2:He with varied pressure. J_{rms} is the rms current density calculated by dividing the discharge current by the electrode plate area, after accounting for a stray capacitance of 23 pF.

Figure 15. E/N of the positive column determined from CVC for 3-cm gap transverse RF discharge flowing 10:33 mmol/s O_2:He with varied pressure.

F. Discharge Scaling Experiments

Scaling ElectriCURL to high laser powers starts with transferring the most power possible into the O_2(a) state in the discharge. High yields of O_2(a) are important, but high flow rates of oxygen and the efficiency of transferring the power into O_2(a) are also critical. Figure 16 demonstrates a way to increase the O_2(a) yield that may not be as useful as it first appears. The O_2(a) yield is plotted versus the input RF power for four cases of varying diluent ratio. The rectangular cross-section tube that has a ¾ inch electrode gap was used with 20 inch long electrodes. The partial pressure of oxygen was held constant at 20 Torr which corresponds to 30 mmol/s while the helium flow rate changed. The flow of NO was approximately constant at 0.2 mmol/s in all cases. As the helium flow increased, the pressure increased resulting in constant discharge residence time. The peak O_2(a) yield improved with increased diluent, but the RF power required to get those yields increased as well. The solid line on the plot simply connects the peaks of the O_2(a) yield versus power curves for the various diluent ratios to indicate that they increase linearly. The power carried by the O_2(a) is calculated from the yield, total oxygen flow rate, and the energy required to create an O_2(a) molecule (0.98 eV), and it is a useful way to evaluate the efficiency of the discharge. The O_2(a) production efficiency is defined as the O_2(a) power divided by the RF power. The percentage of RF power converted to O_2(a) actually drops with diluent ratio although the yield and power stored in O_2(a) increase. Figure 17 illustrates this using the data from the peak of each curve from Fig. 16. The O_2(a) production efficiency drops from 11% in the 1:2, O_2:He case to 9.5% in the 1:8, O_2:He case. However, this effect is convoluted by increased O_2(a) pooling losses that occur at higher pressures of O_2(a), and therefore, the efficiency drop may not be a result of the increased diluent ratio. Despite the decreased efficiency, 13% O_2(a) yield at 66 Torr is a substantial improvement over the 7% yield using the ¾ inch discharge gap shown in Fig. 6. Even the improved performance from the ¾ inch electrode gap discharge does not provide 13% yield at that high pressure. The consequences of higher helium flow rates on the nozzle and laser resonator along with the efficiency decrease would need to be considered before this effect could be usefully implemented.
As discussed previously, the rectangular quartz tubes failed after repeated use in the vacuum system, so new discharge materials must be found that are stronger, but still allow desirable discharge operation. Alumina is a stronger ceramic with better thermal conductivity than quartz and an extremely high melting point. The dielectric constant of alumina is slightly more than double that of quartz, but discharges through both materials operated similarly as Fig. 18 shows. The yield of O$_2$(a) and oxygen atoms is plotted against RF power. The tubes compared in this case are $\frac{3}{4}$ inch ID with 14 inch long electrodes. This length was dictated by the available alumina tube length and explains the yield being somewhat lower than expected based on previous plots and data. The peak O$_2$(a) yield is similar for the tubes, and the alumina tube produces slightly lower oxygen atom levels which is an unexpected benefit. Alumina discharge tubes with rectangular cross-section should allow high flow rate operation with a small electrode gap without worry of the tube fracturing that goes along with quartz.

Another important consideration for scaling to higher oxygen flow rates is the need to apply increased RF power to the discharge. At sufficiently high flow rates, a single RF generator may not output enough power to pump the O$_2$(a). Two sets of electrodes, each with a high voltage and a ground, can easily be applied to a single tube, but the behavior of such a discharge was unknown until the data in Fig. 19 was acquired. Interference or arcing between the two separate discharges in series on one tube was a concern initially, but no problems of that nature were encountered. Two 10 inch electrode sets were placed on the rectangular tube separated by two inches. They were driven with separate matching networks and power supplies, and half of the power was provided to each electrode set. The 20 inch electrodes and the two 10 inch electrodes provided almost exactly the same O$_2$(a) yield for a given input power as the figure shows. This result may prove useful as more RF power must be provided to the primary gas flow.
Figure 18. \( \text{O}_2(a) \) and oxygen atom yields vs. RF Power for quartz and alumina ¼ inch ID tubes. The flow rates were 10:33:0.18 mmol/s, \( \text{O}_2: \text{He:NO} \) at 40 Torr. The electrodes were 14 inches long.

Figure 19. \( \text{O}_2(a) \) yield vs. RF Power using one discharge and two discharges to input the same total power to the rectangular quartz tube. The flow rates were 45:150:0.2 mmol/s, \( \text{O}_2: \text{He:NO} \) at 40 Torr.

After the failure of the edge-welded rectangular quartz tubes, another, more robust, discharge had to be created in order to continue laser testing. A manifold to connect a series of six ¼ inch OD quartz tubes to the laser cavity was created to replace the rectangular tube. Fig. 1 shows the configuration of the tubes as well as the electrodes. Comparisons between the discharge tube configurations are shown in Fig. 20. The \( \text{O}_2(a) \) and oxygen atom yields are plotted versus RF power. Initially, 20 inch electrodes were applied to the series of tubes, but the tubes did not appear to be receiving a uniform distribution of the power. The visible air glow in the tubes was clearly uneven. The measurement of \( \text{O}_2(a) \) yield was slightly lower than the value from the rectangular tube. Experience with the experiments described by Figs. 19 and 10 suggested that in order to combat the non-uniformity problem, a second discharge could be added upstream of the primary 20 inch electrodes. This second discharge used 6 inch electrodes and was powered from a separate matching network and RF generator. A seemingly high power of 2000 W was required on the primer to cause all the discharge tubes in the primary discharge to light evenly. Once this was accomplished the \( \text{O}_2(a) \) yield increased slightly beyond the yield from the rectangular tube as Fig. 20 illustrates. The oxygen atoms for these three cases are also shown as a function RF power. The oxygen atoms are lower in both configurations using the six circular tubes compared to the rectangular tube. The larger wall surface area in the circular tubes may result in more atom recombination. As previously stated, the ultimate goal for these RF discharges is to efficiently maximize the amount of power stored in the \( \text{O}_2(a) \) state. Figure 21 shows that both the rectangular tube and the series of circular tubes can provide high power levels stored in \( \text{O}_2(a) \). At approximately 90 mmol/s of oxygen, the power in \( \text{O}_2(a) \) is over 1 kW. This figure was created holding the power per \( \text{O}_2 \) constant at 80 W/mmol/s, the pressure constant at 40 Torr, and the diluent ratio constant at 1:3.33, \( \text{O}_2: \text{He} \). The NO flow rate was approximately 1% of the input oxygen flow rate. This linear increase in power with input oxygen flow rate is an excellent sign for ultimately increasing the system’s laser power since the discharge’s efficiency is essentially constant.
Figure 20. O$_2$(a) and oxygen atom yields vs. RF Power for several discharge configurations. The flow rates were 45:150:0.25 mmol/s, O$_2$:He:NO at 40 Torr in each case.

Figure 21. Power stored in O$_2$(a) vs. input oxygen flow rate for two discharge tube configurations at 40 Torr. The power was 80 W/mmol/s, and the diluent ratio of 1:3.33, O$_2$:He. The NO flow rate was approximately 1% of the input oxygen flow rate.

IV. Concluding Remarks and Future Work

The discharge research discussed in this work has been one of the key factors towards the gain and laser power improvements in the ElectricOIL system throughout the last year, and some of the work provides guidelines for future scaling of the system to even higher output powers. Despite the higher discharge operating pressures, ozone was shown not to be a major player in the gases downstream of the discharge when NO was added to the flow limiting the oxygen atom production. Downstream of the discharge, the ozone concentration was two orders of magnitude smaller than the O$_2$(a). Previous work suggested that in order to effectively produce O$_2$(a) at pressures in the range of 30 to 50 Torr, the discharge gap needed to be decreased. An electrode gap of ¾ inch provided many useful results, but as the device is scaled to higher flow rates, the discharge pressure may need to be higher. Data show that at about 50 Torr an even smaller electrode gap of ½ inch should provide higher O$_2$(a) yields. Within the discharge, the O$_2$(a) takes some time to reach equilibrium, and this effect appears to happen faster than the computer modeling predicts. Current discharge configurations do not see a benefit to the O$_2$(a) beyond 20 inches of length or about 5 ms of discharge time. Measurements of E/N using microwave interferometry and estimates from the discharge current and voltage agree reasonably well and will provide guidance for future discharge iterations. Diluent ratio studies show that while the yield increases with helium diluent, the efficiency of O$_2$(a) production slightly decreases, so that method for raising the O$_2$(a) yield may or may not be useful for scaling the system. Alumina was shown to be a convenient material for future high flow rate, small electrode gap discharges. Several of these advances led to extracting 102 W of laser power from the system, and decreased electrode gap, rectangular discharges made from alumina should provide large O$_2$(a) powers that will provide even greater laser output. Over 1 kW of power stored in O$_2$(a) has now been demonstrated for two different discharge configurations.

Acknowledgments

This work was supported by DARPA prime contract HR0011-07-C-0054 on a subcontract from Physical Sciences Inc. (PSI). Additionally, J. Zimmerman and B. Woodard were supported in part by the Directed Energy Professional Society (DEPS) Scholarship Program. The authors gratefully acknowledge the contributions of: W.T. Rawlins, S.J. Davis, and S. Lee (PSI); M. Heaven (Emory Univ.); G.W. Sutton (SPARTA/Cobham Analytic Solutions); J. Mangano (DARPA); and R. Snead (U.S. Army SMDC).

The views, opinions, and/or findings contained in this article are those of the authors and should not be interpreted as representing the official views or policies, either expressed or implied, of the Defense Advanced Research Projects Agency or the Department of Defense. Distribution Statement A (Approved for Public Release, Distribution Unlimited).
References


