Electrodynamic modeling of the ElectriCOIL system

Joseph W. Zimmerman[a], Lawrence W. Skorski[a], Wayne C. Solomon[*a], Mark J. Kushner[b],
Joseph T. Verdeyen[c] and David L. Carroll[**c]
[a]Univ. of Illinois, Dept. of Aeronautical and Astronautical Engineering, Urbana, Illinois 61801
[b]Univ. of Illinois, Dept. of Electrical and Computer Engineering, Urbana, Illinois 61801
[c]CU Aerospace, Urbana, Illinois 61801

ABSTRACT

Modeling studies have shown that fractions of $O_2(^1\Delta)$ may be produced in an electrical discharge that will enable oscillation of a chemical oxygen-iodine laser system in conjunction with injection of pre-dissociated iodine. Results of those studies along with recent experimental results indicate that generation of $O_2(^1\Delta)$ can be optimized by the addition of flow diluents and select choice of process parameters. The model predicts the experimentally observed spatial decay of $O_2(^1\Sigma)$ and shows reasonable agreement with experimentally observed temperatures.

Keywords: ElectriCOIL, COIL, chemical oxygen iodine laser, electric discharge modeling

1. INTRODUCTION

The classical chemical oxygen-iodine laser[1] (COIL) operates on the electronic transition of the iodine atom at 1315 nm, $I(^2P_{1/2}) > I(^2P_{3/2}) + h\nu$. The population inversion is obtained by a near resonant energy transfer between the excited $O_2(^1\Delta)$ molecule and the iodine ground state atom $I(^2P_{3/2})$ via the pumping reaction

$$O_2(^1\Delta) + I(^2P_{3/2}) <=> O_2(^3\Sigma) + I(^2P_{1/2}). \tag{1}$$

In the classical COIL, the $O_2(^1\Delta)$ is produced by a liquid chemistry singlet oxygen generator (SOG). However, the difficulties of employing the liquid SOG system in a range of applications have motivated investigations of iodine excitation using all gas phase methods. A recent example is the demonstration of an all gas phase iodine laser pumped by $NCl(^1\Delta)$ (Henshaw[2]). Our research is addressing the scientific and engineering issues associated with an all gas phase SOG, with the goal of obtaining highly efficient $O_2(^1\Delta)$ generation in an electric discharge that will produce a gas phase COIL[3,4,5] (ElectriCOIL).

Researchers have previously shown that significant quantities of $O_2(^1\Delta)$ can be produced in an electric discharge. $O_2(^1\Delta)$ yield is defined as the ratio of the $O_2(^1\Delta)$ concentration to the concentration of all oxygen species in the flow. Benard and Pchelkin[6] reported 11% yield using a microwave discharge. Fujii[7] reported a yield of 17% with a radio-frequency (RF) discharge. Recently, researchers from Fujisaki Electric[8] showed evidence of 21% $O_2(^1\Delta)$ yield using a microwave discharge. Hill[9] reported 16% yield with a controlled-avalanche discharge. Schmiedberger[10] reported 32% yield at low-pressure (0.43 Torr) using an RF discharge. In prior work[5] we obtained an $O_2(^1\Delta)$ yield of ~16% in flowing RF discharge experiments at a pressure of 2 Torr.

In parallel to our experimental work on the ElectriCOIL concept, a computational investigation of the dynamics of ElectriCOIL has been conducted employing the plasma kinetics model GlobalKin[11] (for study of the electric discharge) and Blaze II[12] (for study of the chemical laser dynamics). The goal of this work is the development of a predictive capability for the ElectriCOIL device, and we report on results from the computational investigation here.

[*] wsolomon@uiuc.edu; phone 1-217-244-7646; fax 1-217-244-0720; http://www.aae.uiuc.edu; Univ. of Illinois, 306 Talbot Lab, 104 S. Wright St., Urbana, IL 61801.
[**] cuaerospace@cuaerospace.com; phone 1-217-333-8274; fax 1-217-244-7757; http://cuaerospace.com; CU Aerospace, 2004 S. Wright St. Ext., Urbana, IL 61801.

2. DESCRIPTION OF THE MODEL

The plasma kinetics model GlobalKin[11] was originally developed to investigate reactions involved in the remediation of toxic gases. Recently, GlobalKin and its associated kinetics package were modified to model the kinetics and electrodynamics of the ElectriCOIL discharge. The code is a time-dependent plug flow model with axial position derived from time and gas velocity. The rates for reactions involving only neutrals and ions are taken from experimentally-derived temperature-dependent rate coefficients. The rate information for electron impact reactions can be specified either from electron-temperature dependent fits or from an offline Boltzmann solver. With the latter method, the electron energy distribution function is solved for and reaction rates are parameterized as a function of electron-temperature.

Modeling of the RF discharge in GlobalKin is achieved by specifying a power deposition (W/cm^3) over the length of the discharge region. This power deposition is expressed as either power-per-volume as a function of position or derived from current density as a function of position. The latter enables modeling the discharge as a constant current density apparatus which is representative of the capacitive and inductive discharges used in the experimental ElectriCOIL system. For a given discharge configuration and power, the results for the two methods are similar in terms of $O_2(^1\Delta)$ yield and temperature, exhibiting only slight differences in the their spatial distributions. For this reason, the discharge is modeled using constant power deposition for the studies presented here. The discharge region is modeled as a quasi-DC power source while the experiment uses an alternating current RF power supply. At the pressures and frequencies of interest, the equivalence of the quasi-DC approximation with RF excitation is well known.

3. PREDICTIONS FOR $O_2(^1\Delta)$ GENERATION

3.1 Results of Modeling a Microwave Discharge

In addition to providing support to the ElectriCOIL experiments, we have also modeled experiments by others to provide insight into fundamental processes and to aid in validation. One such experiment was performed by Benard and Pchelkin[6] and consisted of measuring $O_2(^1\Delta)$ concentration in a microwave discharge using EPR spectroscopy. The microwave device was a McCarroll type cavity excited by a 2480-MHz magnetron with 70 W forward and 2 W reflected. The cavity was mounted over a quartz flow tube with 1 cm I.D. flowing pure oxygen at 0.25 mmol/s. The pressure was 0.45 Torr downstream of the discharge in a 7.6 cm I.D. photometric cell. The yield of $O_2(^1\Delta)$ obtained was (11± 0.5) %.

For purposes of modeling the Benard experiment, the flow through the microwave discharge (1 cm I.D.) was taken to be 2 Torr with a discharge length of 1 cm. 50% of the 70 W forward power was absorbed by the flow, resulting in a power deposition of ~45 W/cm^3. Results for $O_2(^1\Delta)$ yield and gas temperature are in Fig. 1. The calculated yield is ~9% at the discharge exit rising to ~13.5% at 5 cm. These values are in reasonable agreement with the EPR measurement (which was made >90 cm downstream). Although the temperature obtained in the experiment was not reported, the calculated peak value of 635 K is believed to be reasonable. The computed yields of excited oxygen and oxygen atoms are given in Fig. 2. These concentrations of $O_2(^1\Sigma)$ and O atoms relative to $O_2(^1\Delta)$ are larger than those obtained when modeling the ElectriCOIL RF discharge due to the higher electron density obtained in the microwave discharge. The microwave discharge achieves electron densities on the order of $1\text{x}10^{12}$ cm^{-3} which is ten times higher than that for ElectriCOIL.

3.2 Trends in the ElectriCOIL System

GlobalKin has been used extensively to model the electrodynamics of $O_2(^1\Delta)$ generation for the ElectriCOIL system. The primary goal is to investigate methods of producing $O_2(^1\Delta)$ in discharges that are favorable for producing positive gain in a laser cavity. The threshold yield Y_{th} of $O_2(^1\Delta)$ required for positive gain as a function of cavity temperature T_c is given by (Hon[13])

$$Y_{th} = \frac{1}{1 + 1.5e^{(401/T_c)}}, \tag{2}$$

where T_c is in degrees K. This relation is derived from the equilibrium of the forward and backward rates of the pumping reaction, Eqn. 1. The significance of Eqn. 2 is that temperature is a critical parameter and must be controlled in order to produce positive gain. The threshold yield expression has been used to evaluate the performance of various discharge configurations.

Investigation of the trends in the ElectriCOIL discharge began with defining a baseline case. The baseline case is a 30-cm long discharge section with a 4.83 cm I.D. flowing 5 mmol/s of pure oxygen at 2 Torr. The discharge was modeled using constant power deposition where the total absorbed power is 300 W. The initial flow and wall temperatures were 300 K. These conditions are representative of a typical ElectriCOIL discharge experiment using an RF generator. Using this configuration as a starting point, computational studies were performed to analyze the effects of power, helium diluent, and cooling on the performance of the discharge.

The yields of oxygen species for the baseline case are shown in Fig. 3 and gas temperature is shown in Fig. 4. The values for yield obtained at the discharge exit were 17% $O_2(^1\Delta)$, 4% $O_2(^1\Sigma)$, 4% O. The peak temperature was 610 K. Assuming that the laser cavity would be placed 10 cm downstream of the discharge exit where the temperature is 600 K, a 25% yield would be required for positive gain. With the laser cavity placed further downstream where the temperature is 400 K, the required $O_2(^1\Delta)$ yield is lowered to ~19%. Given the 21% yield obtained in this region, conditions are only slightly favorable towards positive gain. The computational baseline is therefore theoretically inadequate for feeding the pumping reactions required to produce a high efficiency subsonic laser.

Beginning with the baseline configuration, the power was varied from 100 W to 1.1 kW. The yield and temperature were noted at the discharge exit and 10 cm downstream of the exit, and compared to the required threshold yield. The results of this study are shown in Fig. 5. The values calculated at the discharge exit are below the threshold yield. The yield increases marginally 10 cm downstream with threshold yield being reached at a power level of ~500 W. For the baseline configuration, there is a power level above which the yield as a function of temperature begins to saturate.

The effects of the diluents were also investigated. 20 mmol/s of helium (4:1) were added and the power parameterization repeated extending the maximum value to 1.3 kW. The purpose of adding helium is to lower the (electric field)/(gas number density), E/N, of the discharge to produce more favorable conditions for $O_2(^1\Delta)$ production and to decrease the temperature of the flow. The results indicate that helium dilution produces more favorable conditions than the pure oxygen cases, as shown in Fig. 6. For the maximum power, the temperature at the discharge exit (which is its maximum location) stays below 900 K, well within the operating range of the ElectriCOIL experiment. The yield exceeds threshold 10 cm downstream with power above 300 W. The addition of helium lowers the average E/N in the discharge region from 2.5×10^{-16} V-cm^2 to 1.0×10^{-16} V-cm^2 for the 300 W cases. The E/N distributions through the discharge for the pure oxygen and 4:1 He:O_2 cases at 300 W are shown in Fig. 7. The high value of E/N at the beginning of the discharge is due to initially low electron concentrations.

The other methods that were investigated to improve discharge performance were pre-cooling, wall cooling, and extending the discharge. The 4:1 He:O_2 case was used as a baseline for these studies. Pre-cooling was modeled by lowering the initial temperature to 150 K to simulate running the primary flow through a liquid N_2 bath. Wall cooling consisted of holding the wall of the flow tube after the exit at 150 K simulating a coolant jacket around the tube. A 50 cm discharge section was used for the extended discharge studies, lowering the specific power deposition by 40%. The consequences of these modifications on yield are shown in Fig. 8. Based on values at 10 cm downstream, none of the alternate schemes performed significantly better than the original 30 cm discharge with a helium diluent. However, the wall-cooling case produced higher yields at powers below ~500 W.

4. CONCLUSIONS

The ElectriCOIL system has been computationally investigated using the discharge model GlobalKin. The model predicts the experimentally observed decay of $O_2(^1\Sigma)$ and shows good agreement with observed experimental temperatures. Model predictions for the baseline 300 W, 30-cm discharge, representing an experimental ElectriCOIL configuration, resulted in $O_2(^1\Delta)$ and $O_2(^1\Sigma)$ yields of 17% and 4% respectively and a 610 K peak discharge temperature.

Different configurations for the ElectriCOIL system were investigated to optimize $O_2(^1\Delta)$ yield. These were the addition of helium diluent, cooling of the primary flow, cooling of the post discharge region, and lengthening of the discharge. The addition of helium to the flow gave the most benefit, shifting the yield above the threshold for powers greater than 500 W. We found that gas pre-cooling, reactor wall cooling, and the use of helium diluent may be effective methods for optimizing the $O_2(^1\Delta)$ generation for the ElectriCOIL system.

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FIGURES

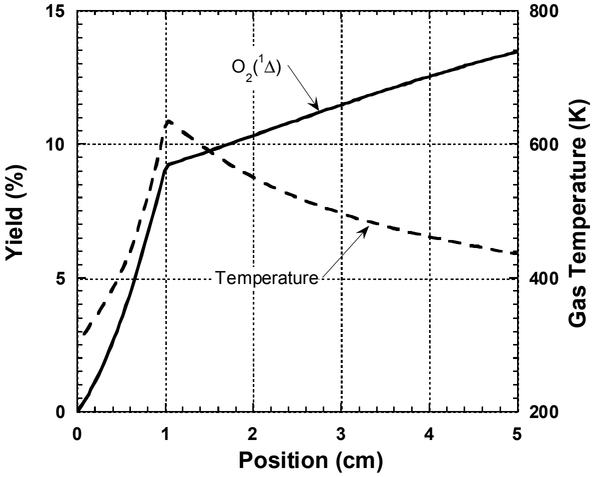

Figure 1: Yield and temperature results modeling the Benard calibration experiment in GlobalKin.

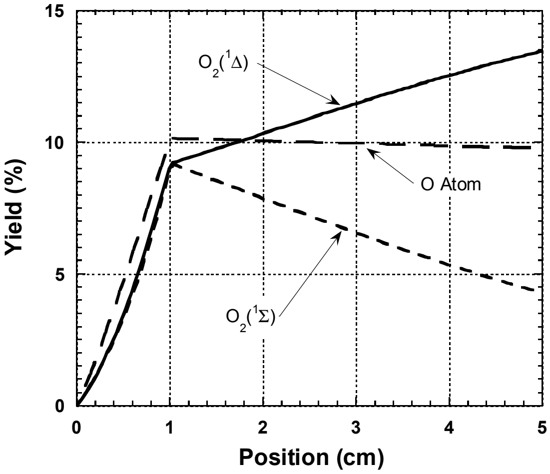

Figure 2: Oxygen species yield results modeling the Benard calibration experiment in GlobalKin.

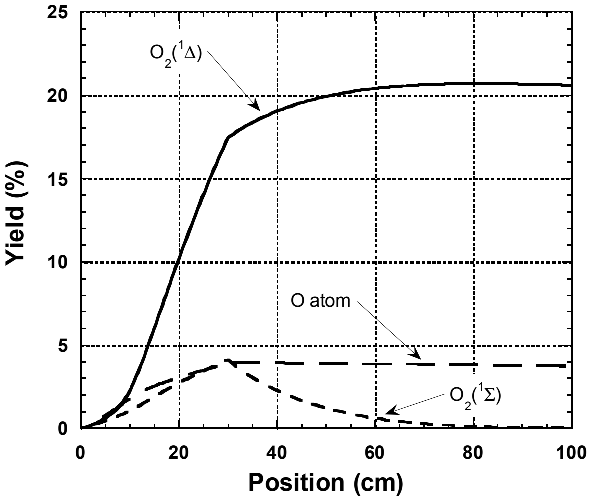

Figure 3: Excited oxygen and oxygen atom yields for the baseline case. 5 mmol/s of O_2 at 2 Torr in a 30-cm, 300 W discharge.

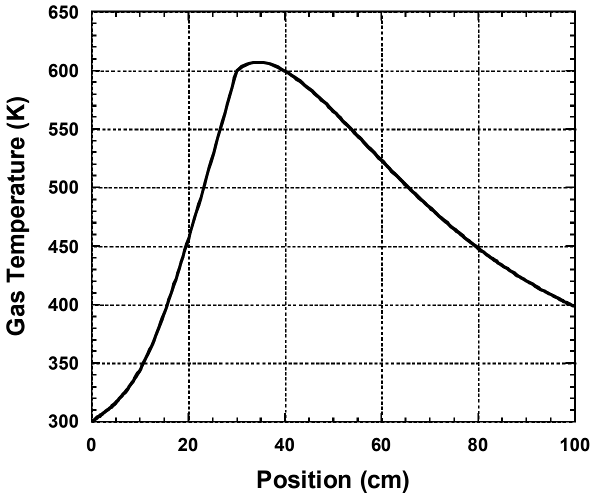

Figure 4: Gas temperature for the baseline case. 5 mmol/s of O_2 at 2 Torr in a 30-cm, 300 W discharge.

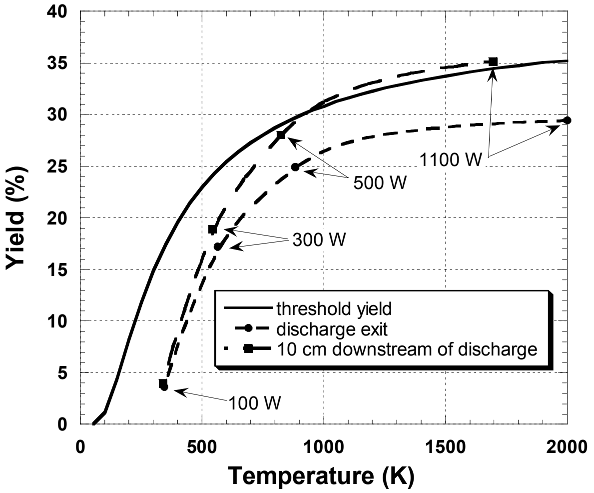

Figure 5: Yield vs. temperature obtained by varying absorbed power compared to threshold yield. 5 mmol/s of O_2 at 2 Torr in a 30-cm discharge.

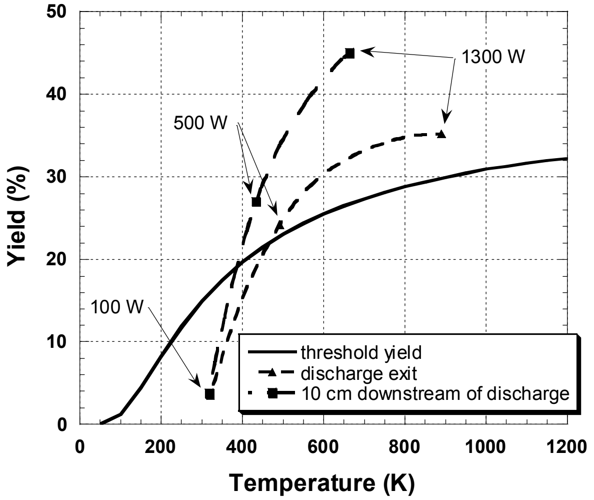

Figure 6: Yield vs. temperature obtained by varying absorbed power compared to threshold yield. 5 mmol/s of O_2 and 20 mmol/s of He at 10 Torr in a 30-cm discharge.

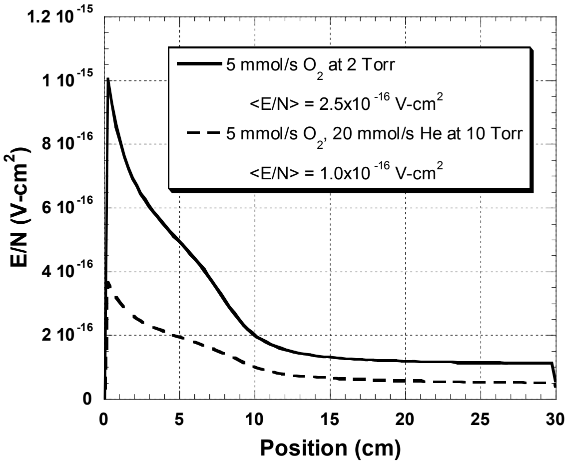

Figure 7: The effect of helium diluent on the E/N distribution. 30-cm discharge at 300 W.

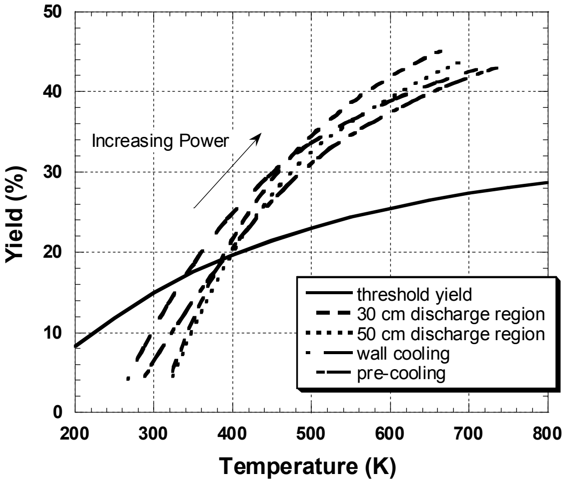

Figure 8: Yield vs. temperature (10 cm downstream of discharge exit) obtained by varying absorbed power compared to threshold yield using different discharge schemes. 5 mmol/s of O_2 and 20 mmol/s of He at 10 Torr in a 30-cm discharge.

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