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Abstract

Theoretical studies have indicated that fractions of $O_2(^1\Delta)$ may be produced in an electrical discharge that may permit lasing of an electric discharge oxygen-iodine laser (ElectriCOIL) system, possibly in conjunction with the injection of pre-dissociated iodine. Results of those studies along with more recent experimental results show that electric excitation is a very complicated process, prompting further investigation with advanced diagnostics along with computational modeling. The electrodynamics and gas kinetics model GlobalKin, including a revised kinetics package was utilized to model the electric discharge region for different gas mixtures and diluents. Results have indicated that performance of the discharge can be increased through varied discharge characteristics and flow diluents including Helium and Argon. The new model predicts a favorable increase in $O_2(^1\Delta)$ yield and reduction of gas temperature with the addition of diluent. The effects of residence time in the discharge region were examined. GlobalKin output was used as input for Blaze II laser modeling to guide future experiments towards a laser demonstration.

1.0 Introduction

The classic chemical oxygen-iodine laser (COIL) [McDermott, 1978] operates on the electronic transition of the iodine atom at 1315 nm, $I^*(^2P_{1/2}) \rightarrow I(^2P_{3/2}) + h\nu$. 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

$$O_2(^1\Delta) + I(^2P_{3/2}) \Leftrightarrow O_2(^3\Sigma) + I(^2P_{1/2}). \qquad (1)$$

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, 2000]. 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 (ElectriCOIL) [Carroll, 2001; King, 2001; Verdeyen, 2002].

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 [Benard, 1978] reported 11% yield using a microwave discharge. Fujii [Fujii, 1994] reported a yield of 17% with a radio-frequency (RF) discharge. More recently, Savin [Savin, 2002] reported 23% $O_2(^1\Delta)$ in a plasmatron microwave discharge. Hill [Hill, 2001] reported 16% yield with a controlled-avalanche discharge. Schmiedberger [Schmiedberger, 2001] reported 32% yield at low-pressure (0.43 Torr) using an RF discharge.

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 [Dorai, 2001; Zimmerman, 2003] (for study of the electric discharge) and Blaze II [Sentman, 1977] (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 herein report on results from the computational investigation.

2.0 Description of the Electrodynamics Model

The plasma kinetics model GlobalKin [Dorai, 2001] 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 [Zimmerman, 2003]. The

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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. 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. At the pressures and frequencies of interest, the equivalence of the quasi-DC approximation with RF excitation is well known.

Zimmerman *et al* [Zimmerman, 2003] showed that GlobalKin-v.4 modeled the microwave discharge data of Benard and Pchelkin [Benard, 1978] reasonably well along with good agreement with existing ElectriCOIL data. Further modeling of the ElectriCOIL discharge was performed to compare with more recent experimental data [Carroll, 2003; Solomon, 2003]. These calculations were performed for both pure oxygen, a 4:1 mix of Helium:Oxygen [Carroll, 2003] and a 3:1 mix of Argon:Oxygen [Solomon, 2003]. In all cases, the partial pressure of oxygen was held fixed at 2.0 Torr. While there are some differences between the experiments and predicted values, given the model's 1-dimensional simplicity it is overall in reasonable agreement with the data. These results give us further confidence that the model is providing an understanding of the discharge physics and that it is making useful predictions that are guiding the experiments.

3.0 GlobalKin Predictions for the ElectriCOIL System as a Function of Diluent

Predictions from a Boltzmann Equation solver that tracks the fraction of electrical power utilized for each electron energy loss process are shown in Figure 1 as a function of different gas mixtures. Shown here is the fraction used to excite the $O_2(^1\Delta)$ state (0.977 eV/exc.) for pure O_2 and mixtures of O_2 with Helium and Argon. In the pure O_2 case nearly 50% of the electrical power can be used to produce $O_2(^1\Delta)$ molecules at an E/N of about 9x10^{-17} volt-cm^2. In the 4:1 He:O_2 case roughly 40% of the electrical power can be used to produce $O_2(^1\Delta)$ molecules at an E/N of about 5x10^{-17} volt-cm^2. In the 3:1 Ar:O_2 case about 60% of the electrical power can be used to produce $O_2(^1\Delta)$ molecules at an E/N of about 4x10^{-17} volt-cm^2. Elastic losses to low atomic weight Helium rob power to $O_2(^1\Delta)$, whereas Argon is beneficial because it has a higher atomic weight and hence its elastic losses are lower. From Fig. 1 it is clear that Argon diluent provides a significant benefit if the E/N can be lowered to the optimal level. Note that Napartovich *et al* [Napartovich, 2001] state that the peak for the pure oxygen case is 43% and a 10:1 Ar:O_2 case is 50%; the reason for this discrepancy is not presently known.

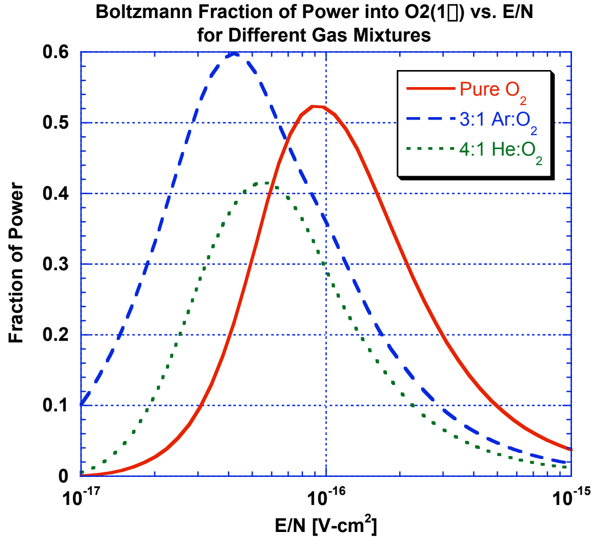

Figure 1: Results of Bolztmann calculation for pure O_2,a 3:1 mix of Ar:O_2,and a 4:1 mix of He:O_2.

Obviously, the efficiency and yield of $O_2(^1\Delta)$ molecules are critically dependent on the field (E) to neutral gas density N, or E/N, with the dependence of the use of electrical power shown in Figure 1. Although these graphs show a clear maximum at E/N ~ 10^{-16} V-cm^2 for pure oxygen, there is no guarantee that the discharge will, in fact, operate at that value. Recent experimental measurements of E/N for our capacitive RF discharge work indicates an E/N of around 2.5x10^{-16} V-cm^2 at 2.0 Torr of pure oxygen, around 0.7x10^{-16} V-cm^2 at 2.0 Torr partial pressure of oxygen mixed with 8.0 Torr partial pressure of He, and around 0.8x10^{-16} V-cm^2 at 2.0 Torr partial pressure of oxygen mixed with 6.0 Torr partial pressure of Ar [Carroll, 2003].

Investigation of the trends in the ElectriCOIL discharge began with defining a baseline case [Zimmerman, 2003]. The baseline case is a 30-cm long discharge section. 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 and argon diluent, and cooling on the performance of the discharge. Figures 2 and 3 show the predicted $O_2(^1\Delta)$ yield and flow temperature for pure oxygen and mixtures of oxygen with helium or argon. It is seen that GlobalKin predicts that the He:O_2 mixture provides the lowest temperatures, but that the Ar:O_2 mixture provides higher yields of $O_2(^1\Delta)$. It should be noted that there is a discrepancy here between the model and experimental data -- experiments show much higher yields with helium diluent [Carroll, 2003] by approximately a factor 2, while the model shows a slightly lower yield with helium; the reason for this is presently unknown and is being investigated.

Based upon various modeling results it appeared that it might be beneficial to yield to have an increased residence time of the oxygen within the discharge region of the flow tube. As such an investigation into the effect of residence time on yield and temperature was conducted. Figure 4 shows that yield increases by approximately 1.5% with residence time by simply increasing the discharge length and that the addition of Argon has an even more pronounced effect by increasing the yield to 20% at the exit of the discharge. There is little effect on the discharge exit temperature by simply increasing discharge length to increase residence time, Fig. 5. However, as has been discussed earlier, one can see the dramatic reduction in discharge exit temperature by adding progressively more Argon diluent, Fig. 5. Overall it appears that the diluent itself has more effect upon discharge performance than does the residence time. These effects are continuing to be studied.

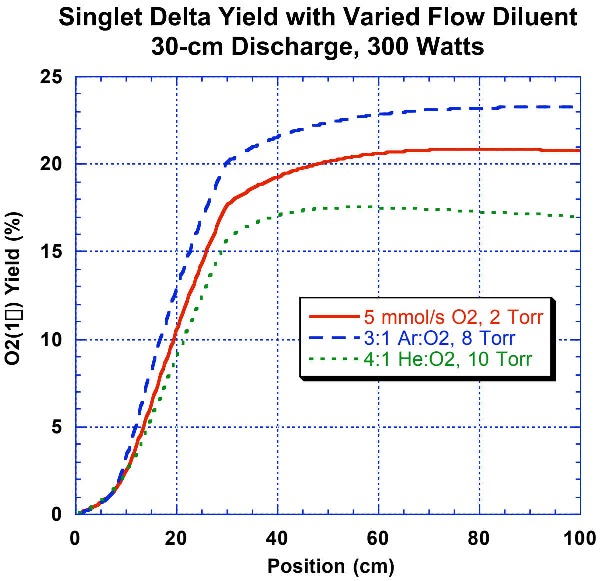

Figure 2. GlobalKin-v.4 predictions of $O_2(^1\Delta)$ yield versus position as a function of gas mixture.

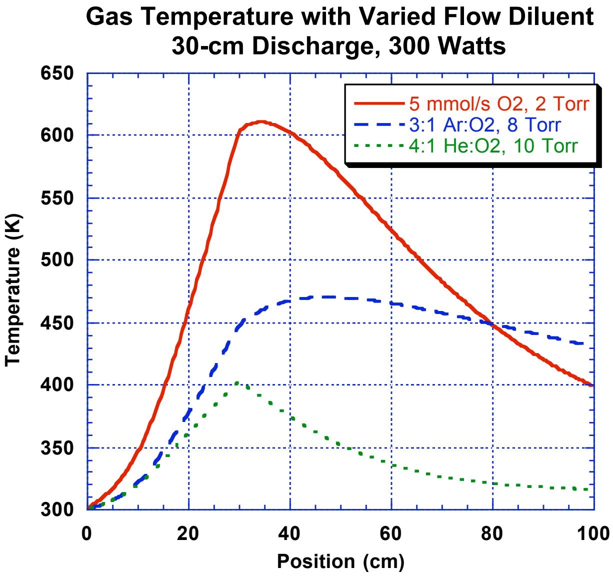

Figure 3. GlobalKin-v.4 predictions of flow temperature versus position as a function of gas mixture.

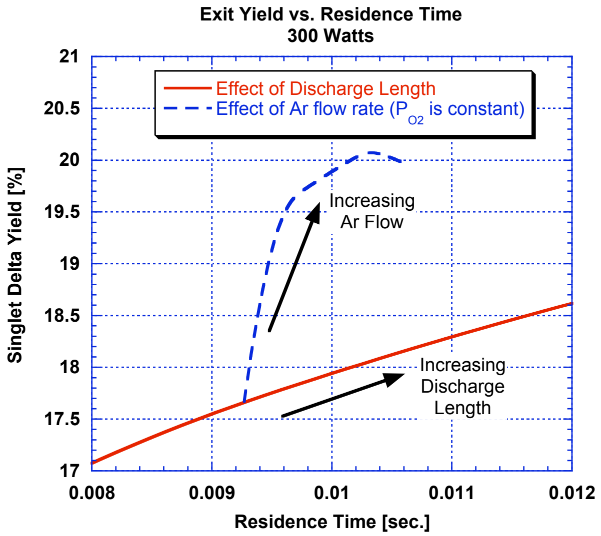

Figure 4: GlobalKin-v.4 predictions of the effect of residence time within the discharge region on singlet-delta yield.

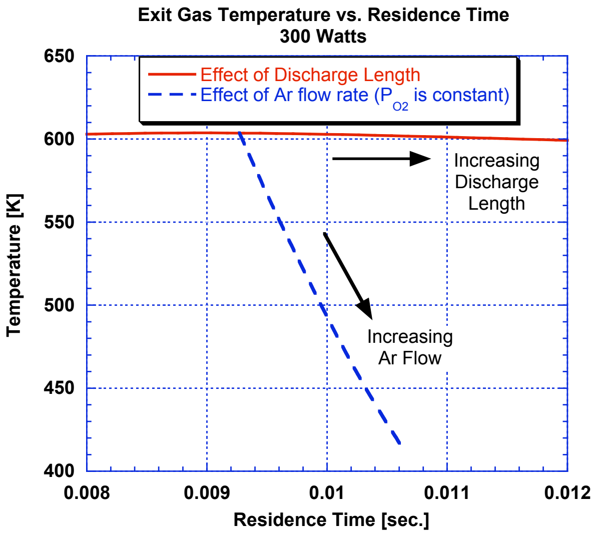

Figure 5: GlobalKin-v.4 predictions of the effect of residence time within the discharge region on temperature at the exit of the discharge.

4.0 Design Experimental Conditions for Lasing

GlobalKin has been used extensively to model the electrodynamics of $O_2(^1\square)$ generation for the ElectriCOIL system. The primary goal is to investigate methods of producing $O_2(^1\square)$ in discharges that are favorable for producing positive gain in a laser cavity. The threshold yield Y_{th} of $O_2(^1\square)$ required for positive gain as a function of cavity temperature T_c is given by [Hon, 1996]

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

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 help evaluate the performance of various discharge conditions and configurations.

A test matrix of flow conditions and rates to be modeled with GlobalKin and Blaze II was put together to determine design points for the lasing experiments. Output from the GlobalKin-v.4 electrodynamics model was used as direct input to the Blaze II laser model. The focus of the matrix study was in a realm of flow conditions and corresponding pressures potentially achievable in laboratory experiments deemed to be most promising based upon relative measurements of singlet-delta levels. Seven different conditions were chosen to model in more detail, Table 1. Note that because of the advantages of lower gas temperatures, mixtures of oxygen with a diluent were examined.

Figures 6 and 7 show subsonic GlobalKin-v.4 predictions of singlet-delta yield versus temperature as a function of absorbed electrical power for these 7 different cases, 10 cm downstream from the exit of the discharge region. Figures 8 and 9 show subsonic GlobalKin predictions of singlet-delta yield versus temperature as a function of absorbed electrical power for these 7 different lab conditions, but 100 cm downstream from the exit of the discharge region. In all four of these figures temperature rises with increasing power levels; the curves start at 0.1 kW and increase to 1.5 kW. Figures 6-9 all start with room temperature gases entering the discharge.

Table 1. Set of flow conditions studied for laser cavity.

Case	O2:He:Ar (mmol/s)	Total Pressure (torr)
1	5:20:0	3.14
2	10:20:10	6.53
3	5:0:15	4.57
4	10:0:20	6.66
5	10:40:0	5.86
6	10:40:10	8.37
7	10:38:2	6.30

A number of important points can be seen from these graphs. First, from Figs. 6 and 7 it is clear that the yield numbers are barely above threshold for the hot gas situation just 10 cm downstream from the exit of the discharge. However, Figs. 8 and 9 show that the situation is dramatically improved by allowing the gas to traverse 100 cm, while cooling, and to permit the conversion of significant amounts of $O_2(^1\square)$ into $O_2(^1\square)$ via reactions with O atoms

[Carroll, 2002a]. Another important point to make is that all of these GlobalKin predictions are for subsonic flow; if the flow is expanded supersonically, then the predicted curves shift to the left as temperature drops. Figures 6-9 are the predictions for a passively cooled flow tube with a wall temperature kept at 300 K; experiments have verified that this passive cooling, which is notably significant, is being predicted with good fidelity [Carroll, 2003; Solomon, 2003]. The most compelling GlobalKin predictions appear to be primarily with Helium diluent because of the significantly lower temperature.

Similar sets of GlobalKin calculations were performed with pre-cooled gas entering the discharge and with a post-discharge cooled flow tube wall (results, not shown). Results with the pre-cooled gas were not substantially different and appear to be more of a second order influence. Results with a post-discharge cooled flow tube wall (set at 150 K) lowered the flow temperatures at the 100 cm point considerably, e.g. the 900 W absorbed power, O_2:He:Ar=10:40:0 mmol/s case without post-cooling had a gas temperature of 354 K at 100 cm, while the same post-cooled case had a gas temperature of 249 K. There were only very minor changes to the species compositions when a post-discharge cooled wall was implemented; the only significant change was the gas temperature.

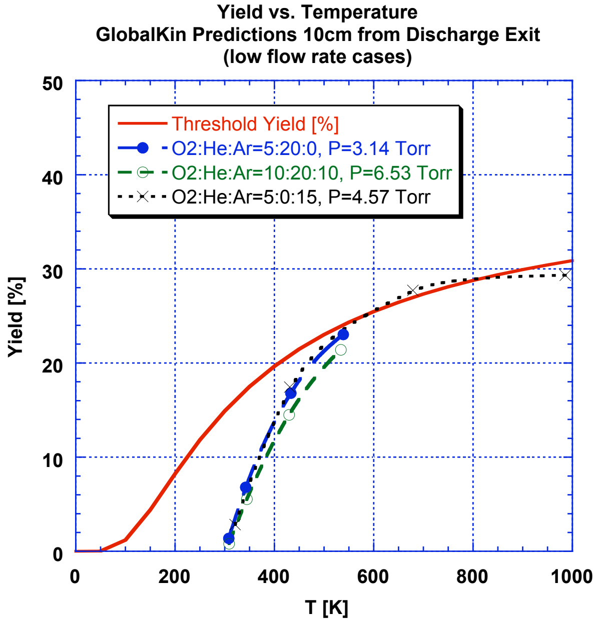

Fig. 6. GlobalKin predictions of singlet-delta yield versus temperature as a function of absorbed electrical power (100, 300, 600 and 900 W) for low flow rate lab conditions, 10 cm downstream from the exit of the discharge region.

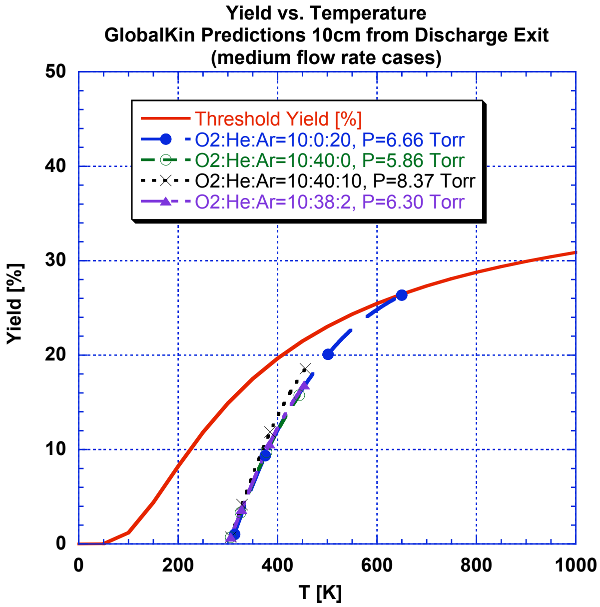

Fig. 7. GlobalKin predictions of singlet-delta yield versus temperature as a function of absorbed electrical power (100, 300, 600 and 900 W) for medium flow rate lab conditions, 10 cm downstream from the exit of the discharge.

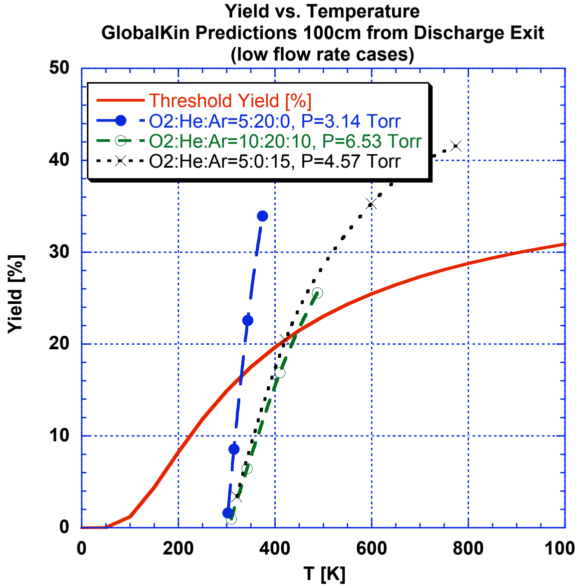

Fig. 8. GlobalKin predictions of singlet-delta yield versus temperature as a function of absorbed electrical power (100, 300, 600 and 900 W) for low flow rate lab conditions, 100 cm downstream from the exit of the discharge region.

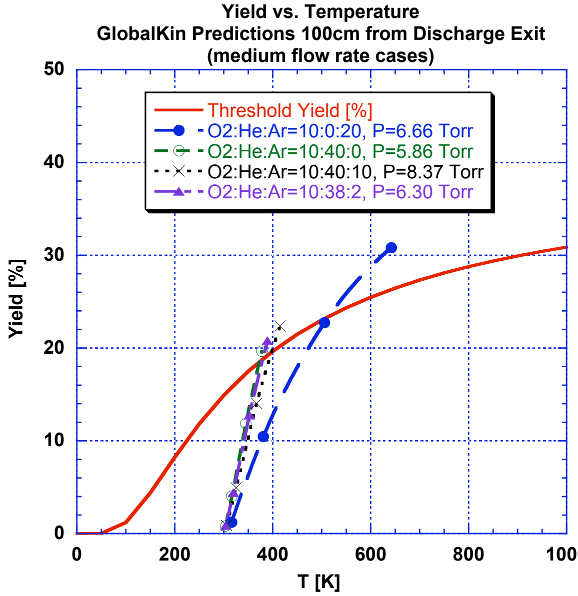

Fig. 9. GlobalKin predictions of singlet-delta yield versus temperature as a function of absorbed electrical power (100, 300, 600 and 900 W) for medium flow rate lab conditions, 100 cm downstream from the exit of the discharge.

For brevity, three representative cases that were modeled with Blaze II will be presented for predictions of gain. These Blaze computations took precise species and temperature output from GlobalKin at the 100 cm point (Figs. 8 and 9), and a throat injected wedge nozzle with a 4.0” gain length was used for the geometry. The wedge nozzle has a 0.20” throat that expands to 1.15” at a distance of 3.60” from the centerline of the injectors. Note that these Blaze calculations have not been corrected for boundary layer formation for this geometry. The ElectriCOIL kinetics package used for these calculations is given in Carroll *et al*, Tables 1 and 2 [Carroll, 2002b]. Figures 10 and 11 show that it is possible to achieve positive gains of 0.03-0.04%/cm for both low (O_2:He:Ar = 5:20:0 mmol/s) and medium (O_2:He:Ar = 10:40:0 mmol/s) flow rate conditions when the absorbed power is 900 W. Positive gains of 0.01-0.02%/cm are achievable when the absorbed power is 600 W.

Positive gain is also attained when Argon is used as the diluent, however, because of the higher temperatures the gain is lower, approximately 0.01%/cm, Fig. 12.

An example of the very significant effect that a post-discharge cooled wall would have on gain is illustrated in Fig. 13. The gain is seen to more than double from 0.04%/cm to 0.08%/cm. This improvement would also show up as an increase in laser power.

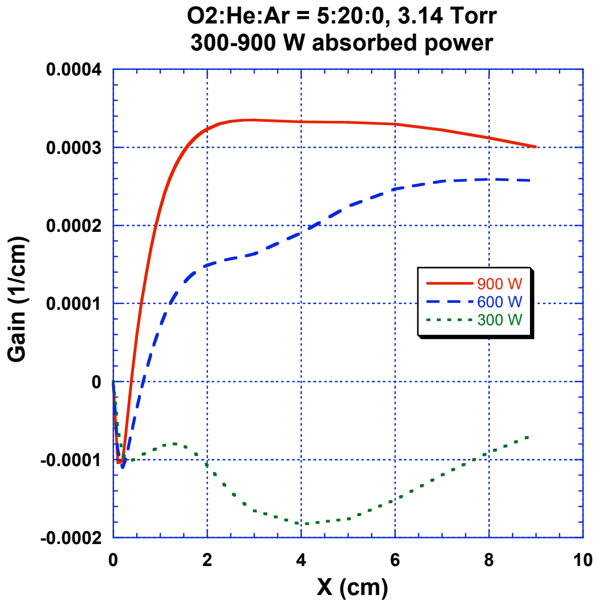
O2:He:Ar = 5:20:0, 3.14 Torr
300-900 W absorbed power

Fig. 10. Blaze II predictions of gain vs. distance from the iodine injection point at the nozzle throat as a function of absorbed electrical power. The secondary flow rates for these cases were 0.10 mmol/s of I_2 and 4.0 mmol/s of He.

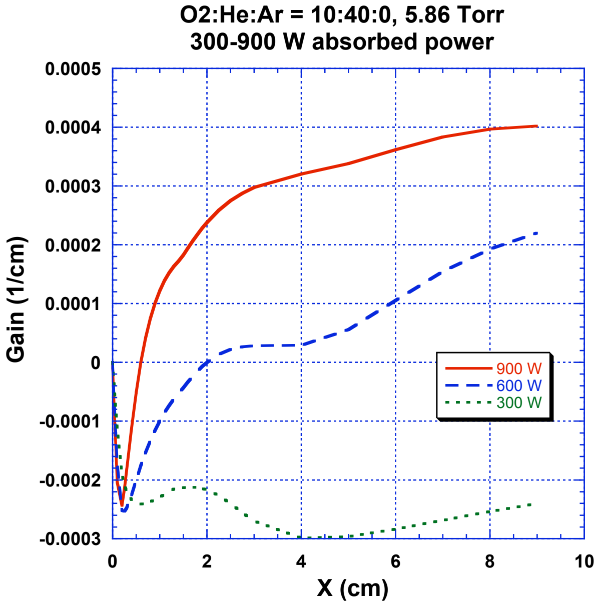
O2:He:Ar = 10:40:0, 5.86 Torr
300-900 W absorbed power

Fig. 11. Blaze II predictions of gain vs. distance from the iodine injection point at the nozzle throat as a function of absorbed electrical power. The secondary flow rates for these cases were 0.20 mmol/s of I_2 and 8.0 mmol/s of He.

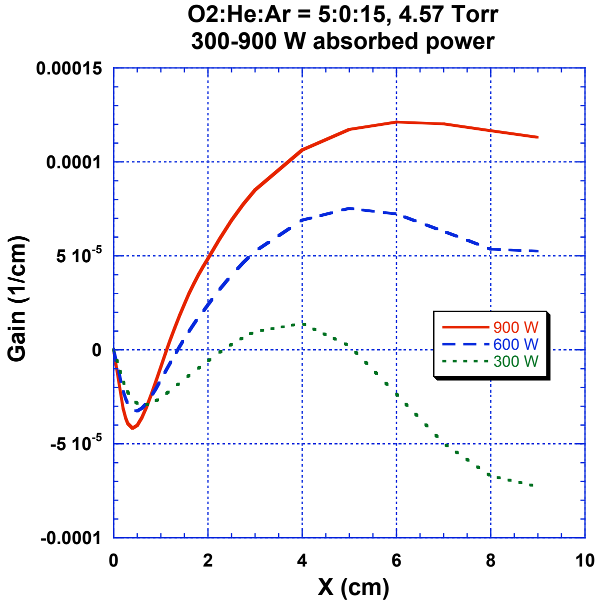
O2:He:Ar = 5:0:15, 4.57 Torr
300-900 W absorbed power

Fig. 12. Blaze II predictions of gain vs. distance from the iodine injection point at the nozzle throat as a function of absorbed electrical power. The secondary flow rates for these cases were 0.10 mmol/s of I_2 and 4.0 mmol/s of He.

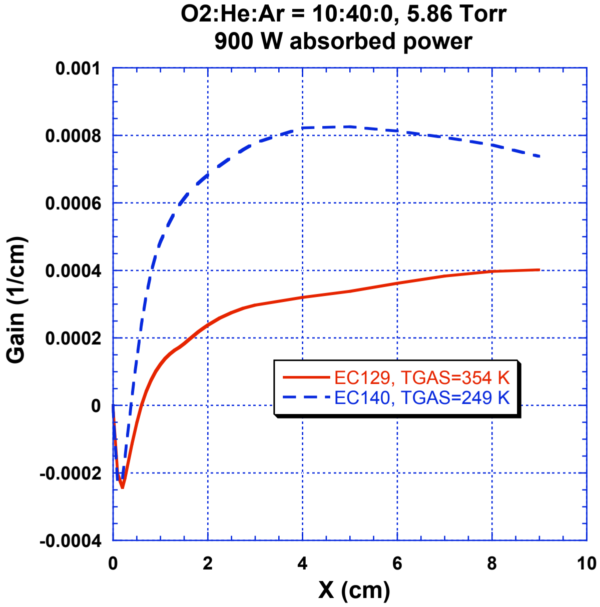
O2:He:Ar = 10:40:0, 5.86 Torr
900 W absorbed power

Fig. 13. Blaze II predictions of gain vs. distance from the iodine injection point at the nozzle throat as a function of gas temperature 100 cm downstream from the discharge. The secondary flow rates for these cases were 0.20 mmol/s of I_2 and 8.0 mmol/s of He.

4.0 Concluding Remarks

One of the objectives of this work was to utilize the electrodynamics and gas kinetics model GlobalKin to model the electric discharge region of an RF discharge in pure oxygen and diluted oxygen mixtures for an electrically assisted chemical iodine laser (ElectriCOIL). Results have indicated that performance of the discharge can be increased through varied discharge characteristics and flow diluents including Helium and Argon. The new model predicts a favorable increase in $O_2(^1\Delta)$ yield and significant lowering of gas temperature when diluent is used in the discharge. The effects of residence time in the discharge region were examined; residence time appears to be of secondary importance when compared to the much more pronounced effects of diluent.

The coupled GlobalKin / Blaze II modeling continues to indicate that it should be possible to achieve positive gain and lasing with a supersonic nozzle. It should be noted that no effort has yet been made to optimize the nozzle, iodine injection point, or secondary flow rates for optimal gain and laser performance. Previous calculations with injection in the subsonic region and a VertiCOIL nozzle were very encouraging [Carroll, 2002b], so we believe this configuration may work well as a possible hardware design improvement. We anticipate that significant improvements can be made as the system continues to evolve.

Acknowledgements

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