Modeling of the ElectriCOIL System

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***Abstract*—Theoretical studies have indicated that sufficient fractions of $O_2(^1\Delta)$ may be produced in an electrical discharge that will permit lasing of an electric discharge oxygen-iodine laser (ElectriCOIL) system. Results of those studies along with more recent experimental results show that electric excitation is a very complicated process that must be investigated with advanced diagnostics along with modeling to better understand this highly complex system. A kinetic package appropriate for the ElectriCOIL system is presented and implemented in the detailed electrodynamic GlobalKin model and the Blaze II chemical laser modeling code. A parametric study with the Blaze II model establishes that it may be possible to attain positive gain in the ElectriCOIL system, perhaps even with subsonic flow. The Blaze II model is in reasonable agreement with early gain data. Temperature is a critical issue, especially in the subsonic cases, and thus it appears that supersonic flow will be important for the ElectriCOIL system. Simulations of a supersonic ElectriCOIL system indicate that it may be possible to attain reasonable performance levels, even at low yield levels of 20% or less. In addition, pre-dissociation of the iodine is shown to be very important for the supersonic flow situation.**

***Index Terms*—Chemical oxygen-iodine laser, chemical oxygen-iodine laser (COIL), discharge oxygen-iodine laser (DOIL) , electric discharge oxygen-iodine laser (ElectriCOIL), radio frequency (RF) excitation of oxygen, singlet-delta oxygen.**

I. INTRODUCTION

THE CLASSIC chemical oxygen-iodine laser (COIL) [30] operates on the electronic transition of the iodine atom at 1315 nm, $I^*(^2P_{1/2}) \rightarrow I(^2P_{3/2}) + h\upsilon$. The population inversion is obtained by a near resonant energy transfer between the excited $O_2(^1\Delta)$ molecule and the I atom ground-state $I(^2P_{3/2})$ via

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

Traditionally, this pumping reaction is fed by a liquid chemistry singlet oxygen generator (SOG). However, the logistic issues of dealing with the liquid SOG systems motivated the investigation of excitation of iodine via all gas phase means by various research groups. AFRL recently demonstrated a new all-gas-phase iodine laser fed by the $NCl(^1\Delta)$ molecule [18]. We believe that it is possible to construct a highly efficient electric generation scheme to provide the precursor energy donor species $O_2(^1\Delta)$ and that an electrically assisted COIL system (ElectriCOIL) [8], [25], [36] can be realized. Researchers at CU Aerospace (CUA) and the University of Illinois at Urbana-Champaign (UIUC) are now addressing the scientific and engineering issues associated with this concept.

Researchers around the world have previously shown that flowing discharge tubes containing ground-state oxygen can produce significant quantities of the desired $O_2(^1\Delta)$ precursor molecules. Benard and Pchelkin [2] reported 11% yield using a low-pressure microwave discharge. Fujii [14] reported good success, 17% yield of $O_2(^1\Delta)$, with a small radio frequency (RF) generator. More recently, workers in Japan [22] from Fujisaki Electric provided some evidence that they could produce 21% $O_2(^1\Delta)$ in a microwave discharge. Hill [20] reported a value of 16% with a controlled-avalanche discharge scheme. Schmiedberger [34] reported a 32% yield under low-pressure conditions (0.43 Torr) with an RF discharge. We have estimated an $O_2(^1\Delta)$ yield of $\approx 16\%$ in our flowing RF discharge experiments at a pressure of 2 Torr [36].

Since the yields of $O_2(^1\Delta)$ using electrical excitation appear to be lower than those with the classic liquid SOG method, it was determined in the original ElectriCOIL concept [8] that atomic iodine injection, rather than molecular iodine injection, will be an important, if not essential, addition to enhancing performance of the ElectriCOIL laser. Experimental work in the area of iodine pre-dissociation has been conducted by Endo *et al.* in Japan [13]. They reported nearly total dissociation from interaction of an Iodine/N_2 stream within the microwave cavity. Iodine pre-dissociation has also been investigated using three-dimensional CFD computations by Madden *et al.* [28]; Madden's results indicated that the injection of atomic iodine slightly downstream of the throat would enhance the power output of a classic COIL device. Recently, CUA and UIUC implemented an LIF experiment that showed 50% dissociation downstream from a dc electric discharge and about 95% in an RF discharge [36].

II. KINETIC MECHANISM

Ongoing ElectriCOIL experiments along with recent GlobalKin [12] simulations have indicated that the kinetic processes taking place in the ElectriCOIL flow system are extremely complex, especially in the discharge region. In addition to the discharge physics, GlobalKin contains an extensive kinetic package including electrons and various positive and negative ions. GlobalKin results indicate the production of atomic oxygen in the discharge region with exit concentrations on the order of those of the $O_2(^1\Delta)$ (Fig. 1). This result is consistent with the experimental measurements by Ivanov *et al.* [23]. GlobalKin results also show that significant quantities of $O_2(^1\Sigma)$ are produced in the discharge along with small, but

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possibly important, levels of O_3. For the baseline case having 5 mmol/s of pure O_2 and 274 W of absorbed power by the flow, the predicted singlet-delta yield is approximately 14% at the exit of the discharge, which rises to higher than 16% in the 4.83-cm I.D. flow tube downstream of the discharge [the rise in yield is primarily a consequence of $O_2(^1\Sigma)$ loss/conversion to $O_2(^1\Delta)$]. The discharge produces approximately 3.6% $O_2(^1\Sigma)$ and 8.8% of O atoms (by mole fraction) at the exit of the discharge.

The GlobalKin model is used to model the discharge physics of the ElectriCOIL system. For the laser system we utilize the Blaze II laser simulation code [35], which contains one-dimensional fluid dynamic equations whose mixing terms were derived from the two-dimensional equations that describe the mixing flowfield in a chemical laser cavity. The Blaze model can be used for premixed, axisymmetric and two-dimensional flows and has proven to be a robust and useful modeling tool for 25 years for several different types of chemical lasers. The use of the Blaze model for COIL simulations is discussed at length by Carroll [5].

In addition to the oxygen kinetics associated with O atoms and O_3, discussions with Heaven [17] suggested the need to incorporate reactions between molecular iodine and atomic oxygen as being an important process. Research by Schmiedberger [34] indicates the use of NO and/or NO_2 as being potentially important for enhancing the production of $O_2(^1\Delta)$. Lastly, there is the desire to perform and model titrations with gases such as NO_2 for O atoms and CO_2 for $O_2(^1\Sigma)$. As such, it is also necessary to expand the classic COIL reaction set that is included in the Blaze II code. Table I presents the classic COIL reaction set used in Blaze II, and Table II presents the expanded mechanism used for the Blaze II modeling of the ElectriCOIL system. It should be noted that the rate constants for reactions 16–20 are significantly different than in the classic COIL reaction set [32]. These new rates are based upon measurements by Lawrence *et al.* [27] of the rotational and vibrational energy transfer rates for selected levels of $I_2(v'' > 20)$. From this data, a numerical model developed by Heaven [15] was used to estimate the global I_2^* deactivation rates listed in Table I [15], [16]. The use of these rates (most significantly reaction 18) was found to provide better agreement with measured gain data than the classic COIL reaction set [6]. These global I_2^* deactivation rates are not appropriate for a newer I_2 dissociation mechanism proposed by Komissarov *et al.* [26]; Komissarov's mechanism is physically more reasonable, but appears to require some more refinement as it significantly underpredicts the iodine dissociation fraction and gain data [4], [29].

Calculations with Blaze II using the added reaction package, listed in Table II, were performed for our flow tube setup with pure oxygen in the flow system. Using output from GlobalKin, a one-to-one comparison was made between the two codes downstream of the discharge and the results were found to be in close agreement, within a few percent (Fig. 2). Thus, it appears that the reduced kinetic package presented in Table II is sufficient for modeling the oxygen kinetics outside of the discharge region.

One of the first things observed in Figs. 1 and 2 is the decay of the $O_2(^1\Sigma)$ concentration with distance downstream. Calculations with the classic COIL kinetic package alone did not show this decay [9]. Examination of the added kinetics pointed clearly to reactions 49 and 50 as being critical to this process and zeroing those reactions eliminated the decay [9]. This is an important observation because it may explain the experimental decay observed by King *et al.* [25] as being attributable to the presence of O atoms in the flow at the exit of the discharge.

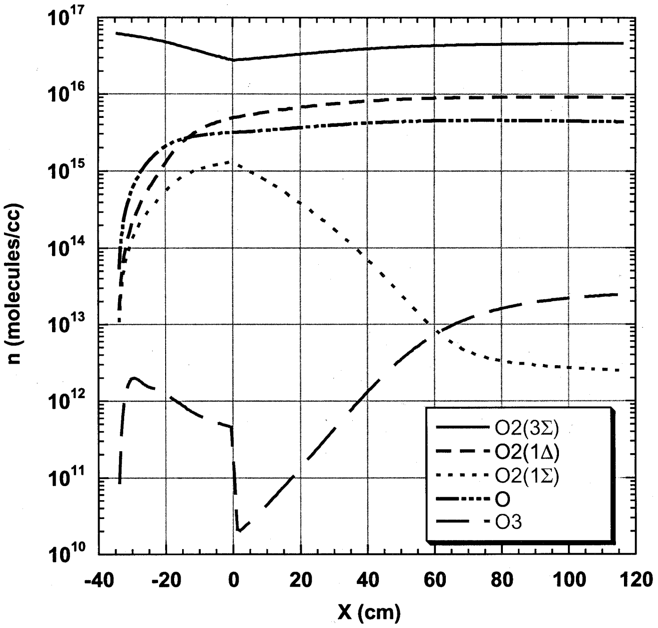

Fig. 1. GlobalKin predictions of oxygen species concentrations as a function of flow distance for a pure oxygen flow at 5 mmol/s and a pressure of 1.94 Torr. The exit of the discharge is at $x = 0$.

III. SUBSONIC STUDIES WITH SINGLE IODINE JET

To better understand the kinetics and flow processes with iodine in the flow, a series of Blaze II computations were run for the flow tube setup with a single iodine jet centered in the subsonic flow tube with the injector hole pointed in the primary flow direction, i.e., parallel to the main flow of oxygen. A schematic of this single jet investigation is illustrated in Fig. 3. While a supersonic flow system will likely produce a higher efficiency laser, this particular subsonic arrangement is very useful for parametric studies.

The baseline condition modeled is a 5 mmol/s primary flow of O_2 and a secondary stream consisting of 0.025 mmol/s of I_2 and 1 mmol/s of secondary He diluent. Based on measurements by Verdeyen *et al.* [36] and many others, the baseline yield of $O_2(^1\Delta)$ is taken to be 16% and $O_2(^1\Sigma)$ is taken as 2% of the flow. Based upon GlobalKin output and experimental data by Ivanov [23], the concentration of O atoms is taken to be the same as $O_2(^1\Delta)$. The pressure of the flow is 1.94 Torr. Thermocouple and spectroscopic temperature measurements of the O_2 flow indicate a temperature of approximately 340 K at the point of iodine injection. The I_2 lines were heated and the secondary flow temperature at the injection point is also assumed to be 340 K. The flow tube has a 1.9" (4.826 cm) I.D. and the iodine injector hole was 3/32" (0.238 cm) in diameter. The initial Mach number of the entire flow (primary plus secondary) for

TABLE I
CLASSIC COIL REACTION SET. NOTE: I REPRESENTS THE $I(^2P_{3/2})$ STATE, I^* THE $I(^2P_{1/2})$ STATE, AND I_2^* IS $I_2(v > 20)$. REACTIONS 16–20 WERE TAKEN FROM [15] AND [16], BASED UPON DATA FROM [27]

k								Rates, cm^3 /molecule-s	Ref.
1	$O_2(^1\Delta)$	+	$O_2(^1\Delta)$	→	$O_2(^1\Sigma)$	+	$O_2(^3\Sigma)$	2.7e-17	Perram, 1988
2	$O_2(^1\Delta)$	+	$O_2(^1\Delta)$	→	$O_2(^3\Sigma)$	+	$O_2(^3\Sigma)$	1.7e-17	Perram, 1988
3	$O_2(^1\Sigma)$	+	$O_2(^3\Sigma)$	→	$O_2(^1\Delta)$	+	$O_2(^3\Sigma)$	3.9e-17	Perram, 1988
4	$O_2(^1\Sigma)$	+	H_2O	→	$O_2(^1\Delta)$	+	H_2O	6.7e-12	Perram, 1988
5	$O_2(^1\Sigma)$	+	Cl_2	→	$O_2(^1\Delta)$	+	Cl_2	2.0e-15	Perram, 1988
6	$O_2(^1\Sigma)$	+	He	→	$O_2(^1\Delta)$	+	He	1.0e-17	Perram, 1988
7	$O_2(^1\Delta)$	+	$O_2(^3\Sigma)$	→	$O_2(^3\Sigma)$	+	$O_2(^3\Sigma)$	1.6e-18	Perram, 1988
8	$O_2(^1\Delta)$	+	H_2O	→	$O_2(^3\Sigma)$	+	H_2O	4.0e-18	Perram, 1988
9	$O_2(^1\Delta)$	+	Cl_2	→	$O_2(^3\Sigma)$	+	Cl_2	6.0e-18	Perram, 1988
10	$O_2(^1\Delta)$	+	He	→	$O_2(^3\Sigma)$	+	He	8.0e-21	Perram, 1988
11	I_2	+	$O_2(^1\Sigma)$	→	2I	+	$O_2(^3\Sigma)$	4.0e-12	Perram, 1988
12	I_2	+	$O_2(^1\Sigma)$	→	I_2	+	$O_2(^3\Sigma)$	1.6e-11	Perram, 1988
13	I_2	+	$O_2(^1\Delta)$	→	I_2*	+	$O_2(^3\Sigma)$	7.0e-15	Perram, 1988
14	I_2	+	I*	→	I	+	I_2*	3.5e-11	Perram, 1988
15	I_2*	+	$O_2(^1\Delta)$	→	2I	+	$O_2(^3\Sigma)$	3.0e-10	Perram, 1988
16	I_2*	+	$O_2(^3\Sigma)$	→	I_2	+	$O_2(^3\Sigma)$	4.9e-12	Heaven, 1995
17	I_2*	+	H_2O	→	I_2	+	H_2O	1.7e-11	Heaven, 1995
18	I_2*	+	He	→	I_2	+	He	9.8e-12	Heaven, 1995
19	I_2*	+	Cl_2	→	I_2	+	Cl_2	6.3e-12	Heaven, 1995
20	I_2*	+	N_2	→	I_2	+	N_2	8.2e-12	Heaven, 1996
21	I	+	$O_2(^1\Delta)$	→	I*	+	$O_2(^3\Sigma)$	7.8e-11	Perram, 1988
22	I*	+	$O_2(^3\Sigma)$	→	I	+	$O_2(^1\Delta)$	1.04e-10*exp(-401.4/T)	Perram, 1988
23	I	+	$O_2(^1\Delta)$	→	I	+	$O_2(^3\Sigma)$	1.0e-15	Perram, 1988
24	I*	+	$O_2(^3\Sigma)$	→	I	+	$O_2(^3\Sigma)$	3.5e-16	Perram, 1988
25	I*	+	$O_2(^1\Delta)$	→	I	+	$O_2(^1\Sigma)$	1.0e-13	Perram, 1988
26	I*	+	$O_2(^1\Delta)$	→	I	+	$O_2(^1\Delta)$	1.1e-13	Perram, 1988
27	I*	+	I	→	I	+	I	1.7e-13	Perram, 1988
28	I*	+	H_2O	→	I	+	H_2O	2.1e-12	Perram, 1988
29	I*	+	He	→	I	+	He	5.0e-18	Perram, 1988
30	I*	+	N_2	→	I	+	N_2	5.0e-17	Deakin, 1972
31	I*	+	Cl_2	→	Cl	+	ICl	5.5e-15	Perram, 1988
32	I*	+	Cl_2	→	I	+	Cl_2	8.0e-15	Perram, 1988
33	I*	+	ICl	→	I_2	+	Cl	1.5e-11	Perram, 1988
34	I_2	+	Cl	→	I	+	ICl	2.0e-10	Perram, 1988
35	Cl	+	ICl	→	I	+	Cl_2	8.0e-12	Perram, 1988
36	2I	+	I_2	→	I_2	+	I_2	3.6e-30	Perram, 1988
37	2I	+	He	→	I_2	+	He	3.8e-33	Busch, 1981
38	2I	+	N_2	→	I_2	+	N_2	4.2e-32	Busch, 1981
39	I* + I	+	I_2	→	I_2(B)	+	I_2	3.6e-30	Perram, 1988
40	I_2(B)			→	2I			1.0e+6	Perram, 1988

these conditions is 0.097. Note that the Blaze II starting point for Figs. 4–11 is at the iodine injection point, which is downstream of the RF discharge.

Experimental data along with the predicted gain curve for the baseline case are shown in Fig. 4. Gain data were taken with a Physical Sciences Inc. Iodine Scan system [10]. It is clear that there is only absorption for the baseline conditions, but that the model is in reasonable agreement with the data. The model predicts an initially fast drop in gain in the mixed region near the iodine injection point ($x = 0$). This is a consequence of the yields being low such that almost all of the entrained excited oxygen species into the mixed region are initially used for the process of dissociating molecular I_2, resulting principally in the production of ground-state I atoms and relatively little I*. As the mixed region grows and larger portions of excited oxygen species are entrained into the mixed flow downstream, the gain subsequently rises. Examining the fluid properties in the simulated flow reveals that the temperature of the mixed flow region

TABLE II
ADDITIONAL ELECTRICOIL RELATED REACTIONS. NOTE THAT THE RATE FOR REACTION 62 IS UNKNOWN (ZERO IS PRESENTLY USED AS THE RATE), BUT THE REACTION COULD POTENTIALLY BE IMPORTANT AND SO IS NOTED IN THE MECHANISM

k								Rates, cm^3 /molecule-s	Ref.
41	2O	+	He	→	$O_2(^3\Sigma)$	+	He	4.5e-34*exp(630/T)	Herron, 2001
42	2O	+	$O_2(^3\Sigma)$	→	$O_2(^3\Sigma)$	+	$O_2(^3\Sigma)$	4.5e-34*exp(630/T)	Herron, 2001
43	2O	+	$O_2(^1\Delta)$	→	$O_2(^3\Sigma)$	+	$O_2(^1\Delta)$	4.5e-34*exp(630/T)	Herron, 2001
44	2O	+	O	→	$O_2(^3\Sigma)$	+	O	4.5e-34*exp(630/T)	Herron, 2001
45	O + $O_2(^3\Sigma)$	+	He	→	O_3	+	He	6.0e-34*$(T/300)^{-2.8}$	Atkinson, 1997
46	O + $O_2(^3\Sigma)$	+	$O_2(^3\Sigma)$	→	O_3	+	$O_2(^3\Sigma)$	6.0e-34*$(T/300)^{-2.8}$	Atkinson, 1997
47	O + $O_2(^3\Sigma)$	+	$O_2(^1\Delta)$	→	O_3	+	$O_2(^1\Delta)$	6.0e-34*$(T/300)^{-2.8}$	Atkinson, 1997
48	O + $O_2(^3\Sigma)$	+	O	→	O_3	+	O	6.0e-34*$(T/300)^{-2.8}$	Atkinson, 1997
49	$O_2(^1\Sigma)$	+	O	→	$O_2(^1\Delta)$	+	O	7.2e-14	Atkinson, 1997
50	$O_2(^1\Sigma)$	+	O	→	$O_2(^3\Sigma)$	+	O	0.8e-14	Atkinson, 1997
51	$O_2(^1\Delta)$	+	O	→	$O_2(^3\Sigma)$	+	O	2.0e-16	Herron, 2001
52	$O_2(^1\Sigma)$	+	O_3	→	$2O_2(^3\Sigma)$	+	O	1.54e-11	Atkinson, 1997
53	$O_2(^1\Sigma)$	+	O_3	→	$O_2(^1\Delta)$	+	O_3	3.3e-12	Atkinson, 1997
54	$O_2(^1\Sigma)$	+	O_3	→	$O_2(^3\Sigma)$	+	O_3	3.3e-12	Atkinson, 1997
55	$O_2(^1\Delta)$	+	O_3	→	$2O_2(^3\Sigma)$	+	O	5.2e-11*exp(-2840/T)	Atkinson, 1997
56	O	+	O_3	→	$O_2(^3\Sigma)$	+	$O_2(^3\Sigma)$	8.0e-12*exp(-2060/T)	Atkinson, 1997
57	$O_2(v)$	+	$O_2(^3\Sigma)$	→	$O_2(^3\Sigma)$	+	$O_2(^3\Sigma)$	4.0e-14	Atkinson, 1997
58	$O_2(v)$	+	He	→	$O_2(^3\Sigma)$	+	He	1.3e-13	Atkinson, 1997
59	I_2	+	O	→	IO	+	I	1.4e-10	Atkinson, 1997
60	IO	+	O	→	I	+	$O_2(^3\Sigma)$	1.5e-10	Payne, 1998
61	I	+	O_3	→	IO	+	$O_2(^3\Sigma)$	2.0e-11*exp(-890/T)	Atkinson, 1997
62	I*	+	O	→	I	+	O	unknown	
63	O	+	NO_2	→	NO	+	$O_2(^3\Sigma)$	6.5e-12*exp(120/T)	Atkinson, 1997
64	O	+	NO	→	NO_2			2.5e-17	Kaufman, 1958
65	O + NO	+	$O_2(^3\Sigma)$	→	NO_2	+	$O_2(^3\Sigma)$	1.0e-31*$(T/300)^{-1.5}$	Atkinson, 1997
66	$O_2(^1\Sigma)$	+	CO_2	→	$O_2(^1\Delta)$	+	CO_2	4.1e-13	Atkinson, 1997

is very high (Fig. 5), which is primarily a consequence of the heat release that occurs during the iodine dissociation process. Temperature data as a function of flow distance were not taken, but a typical measured temperature for these conditions is approximately 500 K; the model is in reasonable agreement with this experimentally measured flame temperature.

Temperature is a critical issue because of the equilibrium of the forward and backward rates of the pumping reaction, (1), [reactions 21 and 22, Table I]. The threshold yield, Y_{th}, of $O_2(^1\Delta)$ required for positive gain as a function of the laser cavity temperature, T_{cav}, can be expressed by [21]

$$Y_{th} = \frac{1}{(1+2k_{eq})} = \frac{1}{\left[1+1.5\exp\left(\frac{401}{T_{\mathrm{cav}}}\right)\right]}. \qquad (2)$$

Equation (2) is illustrated graphically in Fig. 6. From Fig. 6, it is clear that the baseline case yield of 16% requires that the mixed stream temperature stay below approximately 320 K for positive gain. Fig. 5 shows that the mixed flow temperature is reaching approximately 500 K, which from Fig. 6 would require a yield of around 23% for positive gain. Hence, given the predicted temperature rise, it is not surprising that the baseline case exhibits absorption.

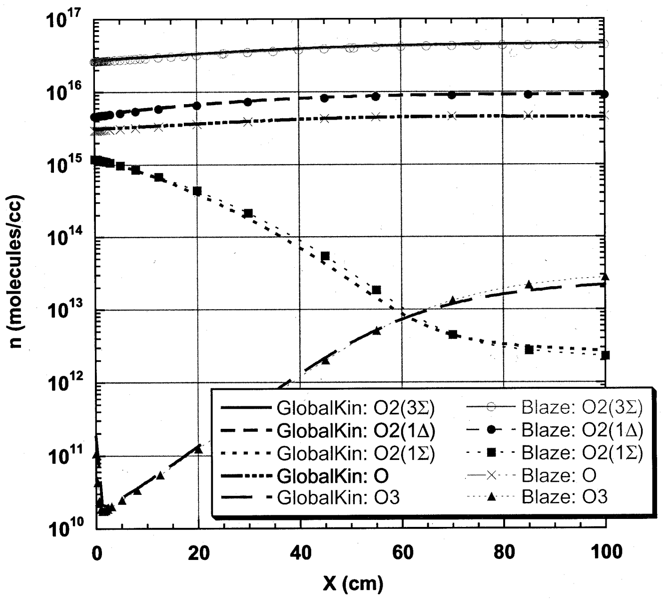

Fig. 2. GlobalKin and Blaze II predictions of oxygen species concentrations as a function of flow distance (downstream of the discharge) for a pure oxygen flow at 5 mmol/s and a pressure of 1.94 Torr.

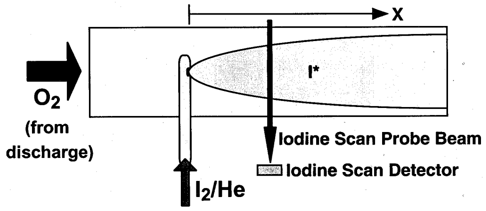

Fig. 3. Schematic of single-jet subsonic experiment and modeling.

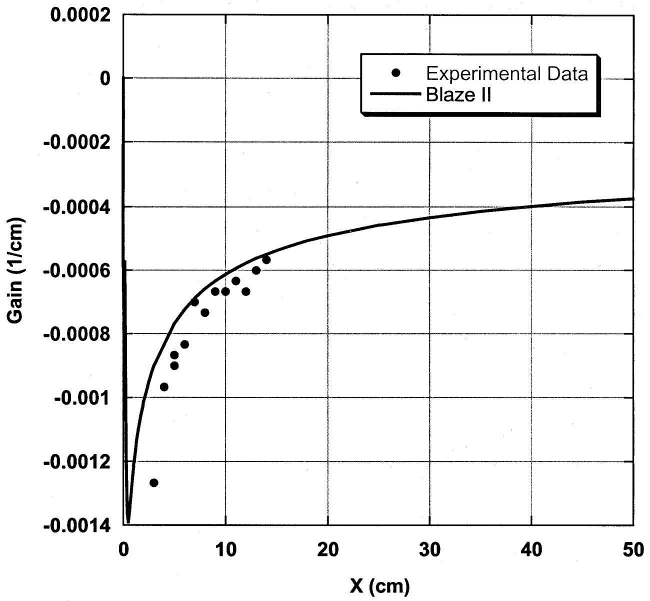

Fig. 4. Gain (absorption) as a function of flow distance for the baseline subsonic mixing case with a single iodine jet. Experimental data are based on an approximate average gain length of 3 cm.

The question that arises is: what routes are there for improving the baseline conditions to produce positive gain from which to perform a lasing demonstration? There are several possible approaches:

1) improve the $O_2(^1\Delta)$ yield from the discharge;
2) pre-cool the primary flow so that the mixed stream temperature will also be lowered;
3) reduce the iodine flow rate to lower the heat released in the I_2 dissociation process;
4) pre-dissociate the iodine molecules prior to injection to eliminate the chemical heat release associated with the I_2 dissociation process;
5) add diluent to absorb chemical heat release and lower the temperature,
6) expand the flow supersonically via a nozzle.

Combinations of these approaches may also be effective in achieving positive gain. As such, a series of parametric calculations were performed to examine the evolution of the gain (or absorption) after the exit of the electric discharge, starting at the iodine injection point (Fig. 3), while varying only one or two parameters at a time. It should be noted that pre-dissociating the molecular iodine prior to injection will eliminate the effective yield loss required for dissociation, however, it is still the mixed stream (cavity) temperature that will dictate the minimum yield required to achieve gain above zero.

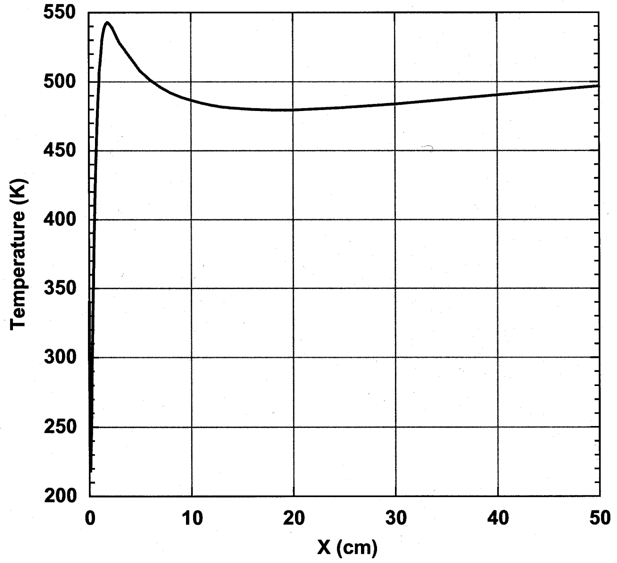

Fig. 5. Mixed stream temperature as a function of flow distance for the baseline subsonic mixing case with a single iodine jet.

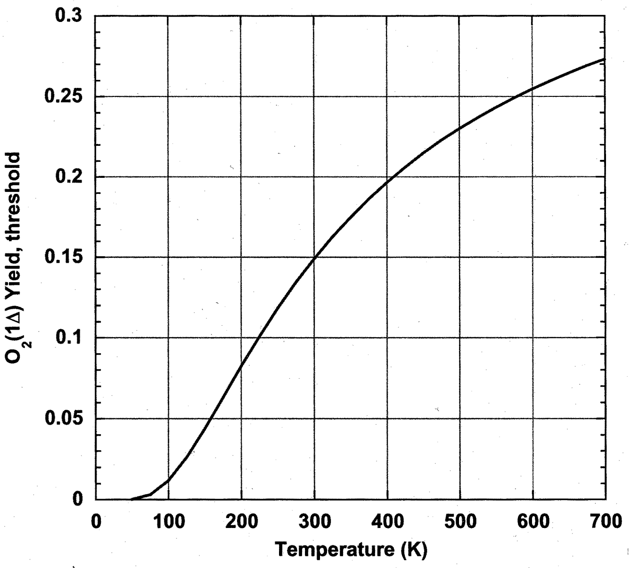

Fig. 6. Threshold yield as a function of laser cavity (mixed stream) temperature for positive gain.

To examine the first possibility a set of cases were run with increasing yield output from the discharge (Fig. 7). With the exception of yield, all the other conditions were the same as for the baseline case illustrated in Figs. 4 and 5 (temperature of 340 K, I_2 flow rate of 0.025 mmol/s). As indicated earlier, it is not surprising that the only one of these calculations producing positive gain was the 25% yield case. There is no question that higher yields from the electric discharge will improve system performance and this will continue to be a key focus of the ongoing experimental program.

A set of calculations [9] was performed for decreasing primary flow temperature. With the exception of the primary flow

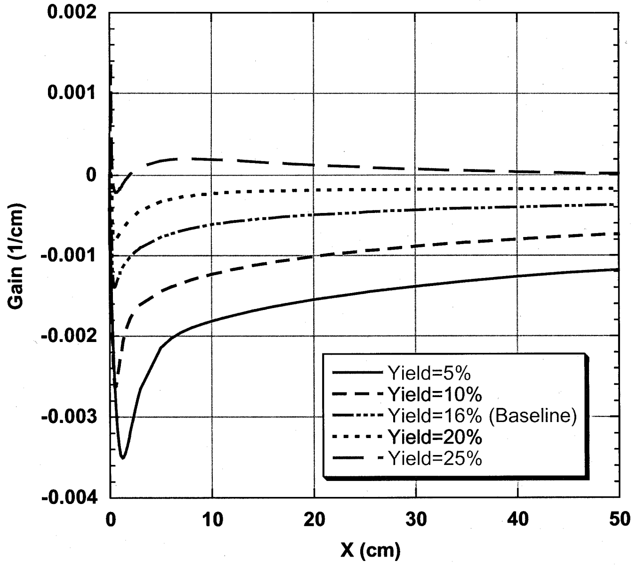

Fig. 7. Gain (absorption) versus flow distance as a function of $O_2(^1\Delta)$ yield for subsonic mixing with a single iodine jet.

temperature, all the other conditions were the same as for the baseline case illustrated in Figs. 4 and 5 (yield of 16%, I_2 flow rate of 0.025 mmol/s) for this second set of calculations. While it was found (results not shown for brevity) that this reduced the amount of absorption (increased the gain), it was also found that the temperature of the mixed stream was still too high (above 320 K for a yield of 16%). The combination of the hot iodine plus the heat released from the chemical dissociation reactions was enough to keep the temperature too high despite a primary flow temperature of as low as 140 K. However, the trend indicated by lowering the primary stream temperature was useful and strongly suggests the possibility of lowering the primary temperature in conjunction with one of the other approaches. As such, calculations were run to examine the combined effects of a lower iodine flow rate of 0.010 mmol/s along with lowered primary flow temperatures (Figs. 8 and 9). With the exception of the primary flow temperature and I_2 flow rate, all the other conditions were the same as for the baseline case illustrated in Figs. 4 and 5 (yield of 16%) for this third set of calculations. Figs. 8 and 9 show that positive gain is predicted for the 140 K primary flow temperature case.

Simulations to test the effects of pre-dissociating the molecular iodine prior to injection were studied next. A calculation with the baseline conditions and 100% iodine pre-dissociation showed a reduced absorption and a significantly reduced temperature, but still above 350 K. As such a set of calculations were performed for the nominal 0.025 mmol/s of I_2, but with a primary flow temperature of 140 K (Figs. 10 and 11). With the exception of the primary flow temperature (fixed at 140 K, rather than 340 K) and iodine dissociation fraction, all the other conditions were the same as for the baseline case illustrated in Figs. 4 and 5 (yield of 16%, I_2 flow rate of 0.025 mmol/s) for this fourth set of calculations. Fig. 11 illustrates how injection of pre-dissociated iodine substantially drops the initial temperature rise in the mixed stream that occurs from the kinetics associated with dissociating the iodine molecules.

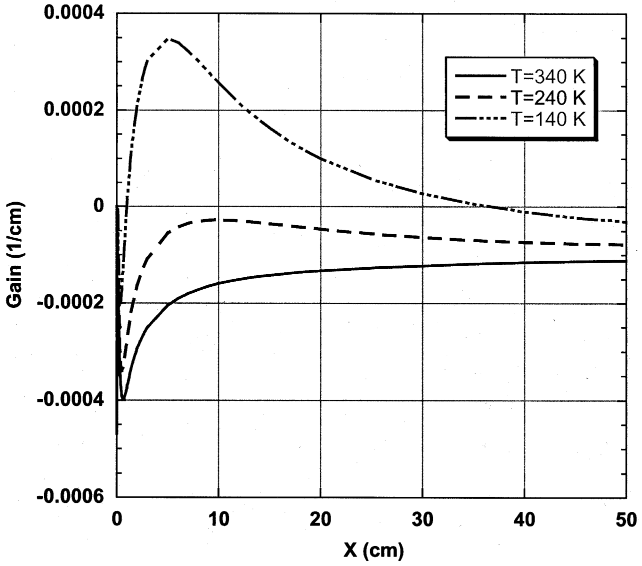

Fig. 8. Gain (absorption) versus flow distance as a function of primary flow temperature for subsonic mixing with a single iodine jet flowing 0.010 mmol/s of I_2.

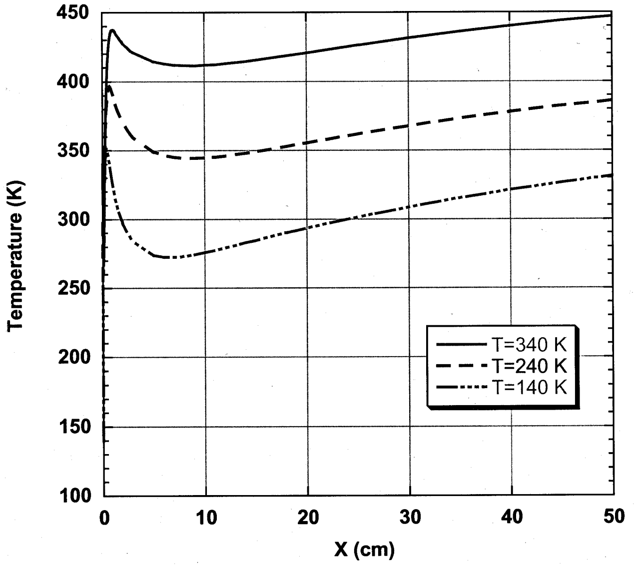

Fig. 9. Mixed stream temperature versus flow distance as a function of primary flow temperature for subsonic mixing with a single iodine jet flowing 0.010 mmol/s of I_2.

Other cases that were run in the parameter space with a fixed yield of 16% that produced positive gain were the 0.010 mmol/s of I_2 flow conditions in conjunction with 140 K primary stream flow and both 50% and 100% pre-dissociation. 0.010 mmol/s of I_2 flow in conjunction with 240 K primary stream flow and 100% pre-dissociation also gave positive gain.

IV. SUPERSONIC MODELING STUDIES

The VertiCOIL [7], [33] nozzle, and typical flow conditions were chosen as the starting point for studies of performance of a supersonic ElectriCOIL system. For these calculations, the iodine is injected into the subsonic region of the flow with the nozzle throat located approximately 1.1-cm downstream from

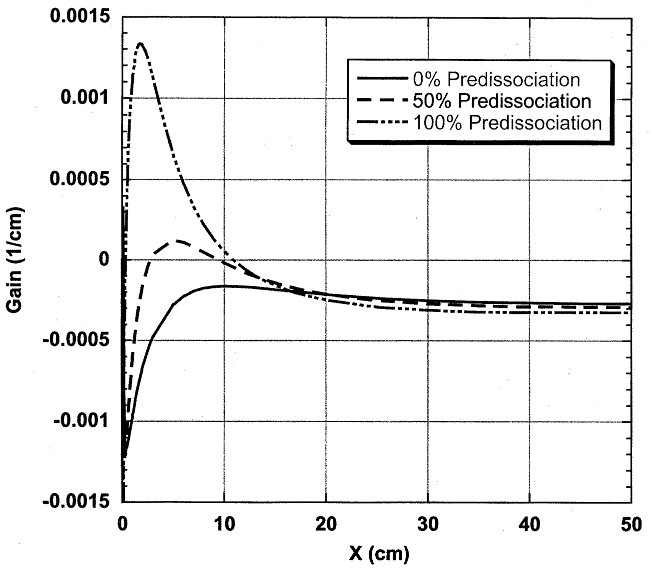

Fig. 10. Gain (absorption) versus flow distance as a function of I_2 pre-dissociation percentage for subsonic mixing with a single iodine jet and primary flow temperature of 140 K.

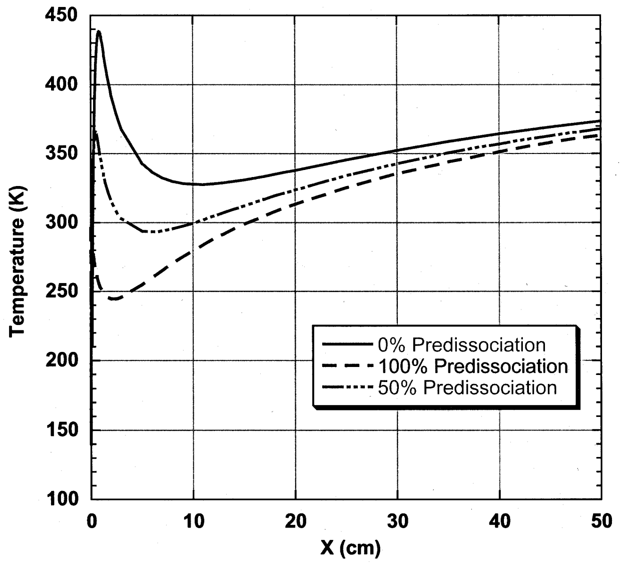

Fig. 11. Mixed stream temperature versus flow distance as a function of I_2 pre-dissociation percentage for subsonic mixing with a single iodine jet and primary flow temperature of 140 K.

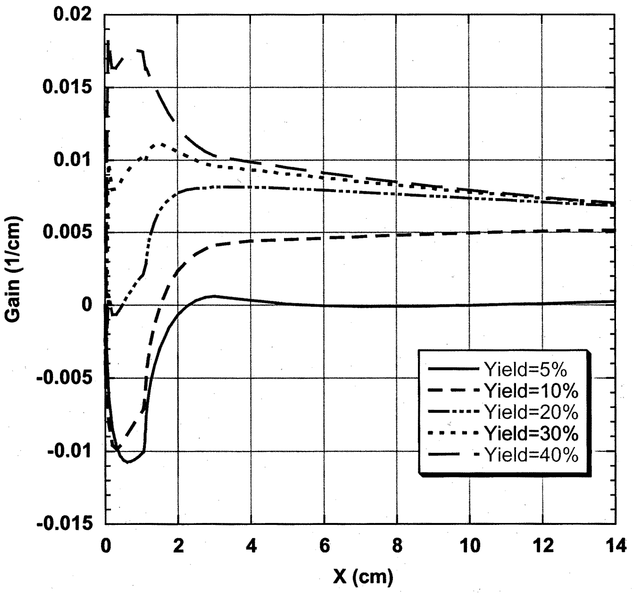

Fig. 12. Gain versus flow distance as a function of yield for supersonic mixing with multiple iodine jets.

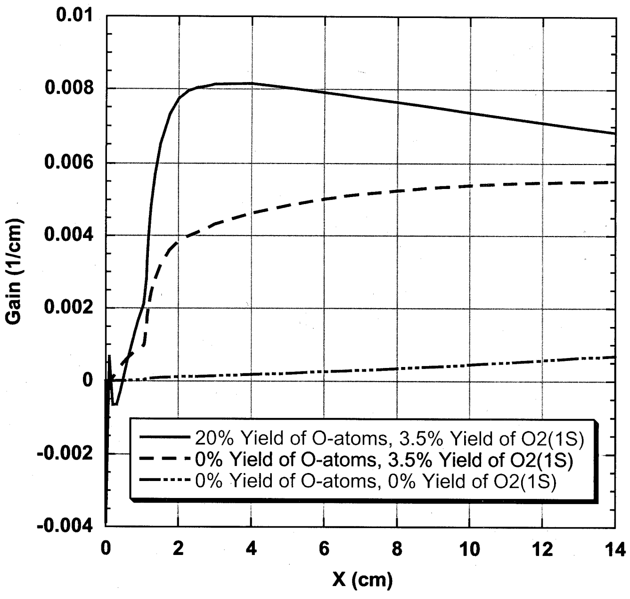

Fig. 13. Gain versus flow distance as a function of O atom and $O_2(^1\Sigma)$ yields for supersonic mixing with multiple iodine jets.

the injection point. The first set of calculations presented here varied the yield of $O_2(^1\Delta)$ to determine what level of yield was necessary to achieve a lasing condition in a supersonic ElectriCOIL system (Fig. 12). Note that these supersonic calculations include an O-atom concentration equal to that of $O_2(^1\Delta)$ and a concentration of $O_2(^1\Sigma)$ equal to 20% that of the $O_2(^1\Delta)$. Of considerable interest is that a yield of only 5% produces optical transparency (zero gain) for a supersonic system, a 10% yield gives a gain of approximately 0.5%/cm, and higher yields provide yet higher gains. These results direct us since they illustrate the point that positive gain should be achievable in a supersonic system because the supersonic expansion cools the flow to cavity temperatures below 150 K.

The next set of calculations was made to determine the effect that the O atoms and $O_2(^1\Sigma)$ were playing in the I_2 dissociation process. These calculations were performed while holding the yield of $O_2(^1\Delta)$ fixed at 20%. Fig. 13 shows that the gain is significantly reduced when O atoms are removed from the flow. When $O_2(^1\Sigma)$ is also removed, the gain becomes very small, but still positive. An examination of the I_2 dissociation fraction (Fig. 14) shows that both O atoms and $O_2(^1\Sigma)$ are playing a very significant role in dissociating the injected molecular iodine in the ElectriCOIL system. Essentially, complete dissociation of I_2 occurs very rapidly in an excess of O atoms and $O_2(^1\Sigma)$, leaving

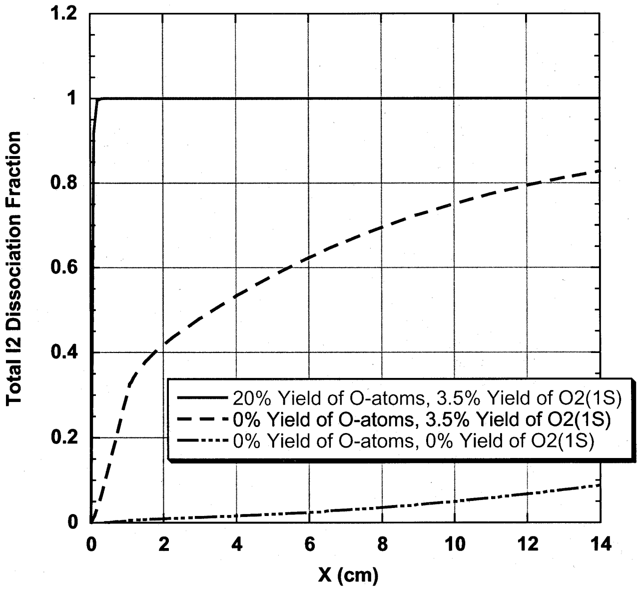

Fig. 14. Iodine dissociation fraction versus flow distance as a function of O atom and $O_2(^1\Sigma)$ yields for supersonic mixing with multiple iodine jets.

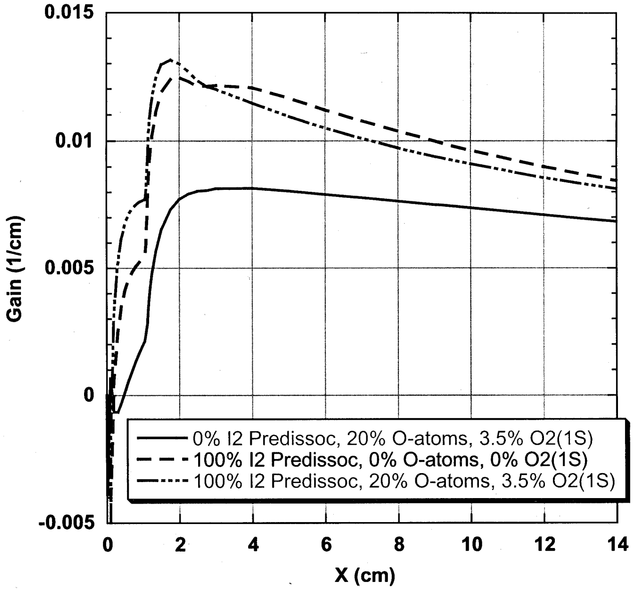

Fig. 15. Gain versus flow distance as a function of I_2 pre-dissociation and O atom and $O_2(^1\Sigma)$ yields for supersonic mixing with multiple iodine jets.

the $O_2(^1\Delta)$ to pump atomic iodine to the lasing level. This is a significant difference from the classic COIL system in which no O atoms are present and $O_2(^1\Sigma)$ only represents a mole fraction of approximately 10^{-5}.

The last set of calculations presented for supersonic flow looks at the effect of pre-dissociating the molecular iodine prior to injecting into the primary oxygen core flow (Fig. 15); these calculations were performed while holding the yield of $O_2(^1\Delta)$ fixed at 20%. When the molecular iodine is completely pre-dissociated, the gain curve, even with a 20% yield, increases from 0.8%/cm to a peak of 1.3%/cm; this compares very favorably with the classic COIL type gain curve. It is also interesting to see that when the O atoms and $O_2(^1\Sigma)$ are removed from the flow while at the same time injecting pre-dissociated I_2, the gain curve is very similar to the case of pre-dissociated I_2 with O atoms and $O_2(^1\Sigma)$; this indicates that the presence of O atoms and $O_2(^1\Sigma)$ do not significantly enhance the gain curve when the I_2 is pre-dissociated prior to injection. It is also important to point out that these predictions indicate it is preferable to pre-dissociate the I_2 than to rely only on the O atoms and $O_2(^1\Sigma)$ for dissociation of molecular iodine in the ElectriCOIL system.

V. CONCLUDING REMARKS

Ongoing ElectriCOIL experiments along with recent GlobalKin simulations have indicated that the kinetic processes taking place in the ElectriCOIL flow system are extremely complex, especially in the discharge region. To properly model the discharge physics and kinetics, GlobalKin contains an extensive kinetic package including electrons and various positive and negative ions. GlobalKin results indicate the production of atomic oxygen in the discharge region with exit concentrations on the order of those of the $O_2(^1\Delta)$ (Fig. 1). This result is consistent with the experimental measurements by Ivanov *et al.* [23]. GlobalKin results also show that significant quantities of $O_2(^1\Sigma)$ are produced in the discharge along with small, but possibly important, levels of O_3. For the baseline case having 5 mmol/s of pure O_2 and 274 W of absorbed power by the flow, the predicted singlet-delta yield is approximately 14% at the exit of the discharge, which rises to higher than 16% in the flow tube downstream of the discharge (the rise in yield is primarily a consequence of singlet-sigma loss/conversion to singlet-delta). The discharge produces approximately 3.6% $O_2(^1\Sigma)$ and 8.8% of O atoms (by mole fraction) at the exit of the discharge.

A modified kinetic package to more accurately simulate the flow of an ElectriCOIL laser system has been implemented in the Blaze II chemical laser model. Using output from GlobalKin, a one-to-one comparison was made between the two codes downstream of the discharge and the results were found to be in close agreement. Thus, it appears that the reduced kinetic package is sufficient for modeling the oxygen kinetics outside of the discharge region. The package predicts the decay in the $O_2(^1\Sigma)$ that has been observed experimentally and suggests that the decay is due to reactions between $O_2(^1\Sigma)$ and O atoms.

Gain data were taken with a Physical Sciences Inc. Iodine Scan system for a subsonic baseline condition having 5 mmol/s primary flow of O_2 and a secondary stream consisting of 0.025 mmol/s of I_2 and 1 mmol/s of secondary He diluent at a pressure of 1.94 Torr. There was only absorption for the baseline conditions. The Blaze II model was used to model this data and was found to be in reasonable agreement with the experimental data.

A parametric study showed that temperature is a critical flow variable in the ElectriCOIL system, especially in the subsonic cases. Two key factors that drive the mixed stream (cavity) flow temperature higher are the chemical kinetics associated with dissociating the iodine molecules, and the deactivation of $O_2(^1\Sigma)$ by O atoms. However, there are several approaches to attaining positive gain.

1) Improve the $O_2(^1\Delta)$ yield from the discharge.
2) Pre-cool the primary flow so that the mixed stream temperature will also be lowered.
3) Reduce the iodine flow rate to lower the heat released in the I_2 dissociation process, thereby lowering the mixed stream (cavity) temperature.
4) Pre-dissociate the iodine molecules prior to injection to eliminate the chemical heat release associated with the I_2 dissociation process.
5) Add diluent to absorb chemical heat release and lower the temperature.
6) Expand the flow supersonically via a nozzle.

Higher discharge yield and supersonic flow expansion individually are enough to produce the necessary lasing conditions, while combinations of the other approaches 2–4 also result in positive gain. Approach 5), the addition of diluent, needs to be investigated in future simulations. This parametric study established that it may be possible to attain positive gain in the ElectriCOIL system, perhaps even with subsonic flow.

Simulations of a supersonic ElectriCOIL system indicate that it may be possible to attain reasonable performance levels, even at low yield levels of 20% or less. The presence of O atoms and $O_2(^1\Sigma)$ in the flow very significantly influence the gain by rapidly dissociating molecular I_2. In addition, pre-dissociation of the iodine is also shown to be very useful for the supersonic flow situation and provides higher gain than without the predissociation; therefore, it is preferable to pre-dissociate the I_2 than to rely only on the O atoms and $O_2(^1\Sigma)$ for dissociation of molecular iodine. Given the critical nature of the temperature issue, it appears that supersonic flow will be required for the ElectriCOIL system to achieve significant performance levels.

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