Kinetics and Scaling of Gain and Lasing in a 1-5 kW Microwave Discharge Oxygen Iodine Laser

Wilson T. Rawlins, Seonkyung Lee, Adam J. Hicks, Ian M. Konen, Emily P. Plumb, David B. Oakes, and Steven J. Davis*

Physical Sciences Inc.
20 New England Business Center
Andover, MA  01810-1077

ABSTRACT

Scaling of Electric Oxygen-Iodine Laser (EOIL) systems to higher powers requires extension of electric discharge powers into the kW range and beyond, with high efficiency and singlet oxygen yield. This paper describes the implementation of a moderate-power (1 to 5 kW) microwave discharge at 30 to 70 Torr pressure in a supersonic flow reactor designed for systematic investigations of the scaling of gain and lasing with power and flow conditions. The 2450 MHz microwave discharge is confined near the flow axis by a swirl flow. The discharge effluent, containing active species including O\textsubscript{2}(a\textsuperscript{1}Σ\textsubscript{g}), O(3P), and O\textsubscript{3}, passes through a 2-D flow duct equipped with a supersonic nozzle and cavity. I\textsubscript{2} is injected upstream of the supersonic nozzle. The apparatus is water-cooled, and is modular to permit a variety of inlet, nozzle, and optical configurations. A comprehensive suite of optical emission and absorption diagnostics monitors the absolute concentrations of O\textsubscript{2}(a), O(3P), O\textsubscript{3}, I\textsubscript{2}, I(2P\textsubscript{3/2}), I(2P\textsubscript{1/2}), small-signal gain, and temperature in both the subsonic and supersonic flow streams. The experimental results include numerous observations of positive gain and lasing in supersonic flow, and the scaling of gain with a variety of flow and reaction rate conditions. The results are compared with kinetics modeling predictions to highlight key discrepancies as well as areas of agreement. The observed gains are generally lower than the predicted values, due in part to chemical kinetics effects and also due to mixing limitations specific to the reagent injection design. We discuss in detail the observed effects related to O-atom chemistry, and their import for scaling the gain to higher levels. We also will present initial beam quality measurements.

Keywords: Oxygen Iodine laser, kinetics, singlet molecular oxygen, beam quality

1. INTRODUCTION

The electric oxygen iodine laser (EOIL) holds considerable promise for a high power gas phase laser with excellent beam quality and good electrical efficiency. We have been using a high power microwave discharge, supersonic flow facility to study the kinetics, small signal gain, and laser output in order to understand the scaling properties of this system. A scaled system has the potential to be a much more compact and lighter-weight alternative to the well known chemical oxygen iodine laser (COIL), developed in 1977\textsuperscript{1} and subsequently scaled to MW levels.\textsuperscript{2} In both EOIL and COIL, electronically excited metastable singlet oxygen, O\textsubscript{2}(a\textsuperscript{1}Δ\textsubscript{g}) [hereafter also referred to as O\textsubscript{2}(a)], collisionally pumps iodine atoms to produce single-line laser output on the I(2P\textsubscript{3/2}→2P\textsubscript{3/2}) transition at 1.315 µm. EOIL offers all the advantages of COIL such as excellent beam quality,\textsuperscript{3} good propagation properties,\textsuperscript{2} and high power operation. However, EOIL eliminates the complex, massive and hazardous liquid-based chemistry system that is required in COIL to produce singlet molecular oxygen to power the laser. At present, EOIL technology test facilities are operated as open-cycle systems. However, due to its all-gas-phase feedstock, the device is well suited for closed-cycle operation. In EOIL test systems to date, flowing He/O\textsubscript{2} mixtures are passed through an electric discharge to produce the O\textsubscript{2}(a).

*davis@psicorp.com; phone 1 978 689-0003; fax 1 978 689-3232; psicorp.com
2. TECHNICAL BACKGROUND

Our technical approach is based on several years of research and development on the chemical kinetics and performance of the electric oxygen iodine laser system.\textsuperscript{4-12} The electric discharge methods employed (high power microwave, radio frequency, and pulser-sustainer) typically operate on mixtures of O\textsubscript{2} and helium and produce several energetic species. Most of these species are short-lived after the gas exits the active discharge. O atoms and O\textsubscript{2}(a) are the primary energetic species that survive into the reaction zone, where I\textsubscript{2} is introduced into the flow stream. A typical system is illustrated in Figure 1 along with the basic mechanisms for excitation of the singlet oxygen, iodine atom, and laser output.

\[ \text{O} + \text{I}_2 \rightarrow \text{IO} + \text{I} \quad \text{I}_2 \text{ dissociation} \]
\[ \text{O} + \text{IO} \rightarrow \text{O}_2 + \text{I} \]
\[ \text{O}_2(a) + \text{I} \rightarrow \text{O}_2(X) + \text{I}^* \quad \text{I}^* \text{ excitation (near-resonant, reversible)} \quad (1) \]
\[ \text{O} + \text{I}^* \rightarrow \text{O} + \text{I} \quad \text{I}^* \text{ quenching} \]
\[ \text{I}^* \rightarrow \text{I} + \text{hv} \quad \text{Lasing} \]

Atomic oxygen provides rapid dissociation of I\textsubscript{2}, so that no O\textsubscript{2}(a) is lost in the dissociation step. However, atomic oxygen in large concentrations is also a significant quencher of the laser emission, so its concentration must be actively controlled. This is done by addition of NO\textsubscript{2} or NO, using the coupled reactions

\[ \text{O} + \text{NO} + \text{M} \rightarrow \text{NO}_2 + \text{M} \]
\[ \text{O} + \text{NO}_2 \rightarrow \text{NO} + \text{O}_2 \]

(2)

to reduce the O concentration (where M signifies a generic third-body collision partner, e.g. O\textsubscript{2} and He). We have previously demonstrated that when the O concentration is sufficiently reduced through reaction with NO\textsubscript{2} or NO, the optical gain is greatly increased and is sufficient to support lasing.\textsuperscript{4-12}

In the reaction scheme given above, when the I\textsuperscript{*}+O quenching reaction rate is sufficiently reduced, the inversion ratio [I\textsuperscript{*}]/[I] is controlled primarily by the reversible energy transfer reaction between O\textsubscript{2}(a) and atomic iodine, Reaction (1). Thus in the absence of other loss terms, the optical gain is directly related to the O\textsubscript{2}(a) yield (i.e. the fraction of the incoming oxygen that is excited to the O\textsubscript{2}(a) state) through the equilibrium constant for Reaction (1). Since the energy of O\textsubscript{2}(a) is slightly higher than that of I\textsuperscript{*}, this equilibrium shifts strongly towards I\textsuperscript{*} excitation as the temperature is reduced. For this reason, test systems usually employ a supersonic nozzle to reduce the temperature of the reacting mixture to 100-200 K. For a supersonically cooled flow of ~200 K, the threshold for optical gain is a singlet oxygen yield of ~8%.

Figure 1. Diagram of an open-cycle EOIL. Several regions of the flow are indicated along with the dominant chemical reactions.
3. ELECTRIC DISCHARGE PHENOMENOLOGY

O$_2$(a) can be generated in a variety of electric discharges and configurations including self-sustained (microwave and RF) and non self-sustained (pulsar-sustainer). The self-sustained microwave and RF methods have produced the highest reported yields and highest output powers.

We have previously published model calculations of excitation, dissociation, and ionization rates in O$_2$/Ar discharges. We perform these calculations with a computer code which solves the Boltzmann transport equation. The code treats all the inelastic processes occurring in the active discharge to evaluate the steady-state electron energy distributions and reaction rate coefficients as functions of E/N and O$_2$ mole fraction in Ar or He. The electron-impact cross section data are taken from the data base discussed extensively by Phelps and co-workers.

Two experimental controls on the electron energy distribution are the E/N of the discharge and the relative amounts of O$_2$ and rare gas (He or Ar) in the gas mixture. E/N is the ratio of the electric field strength E, which is governed by the applied power and discharge geometry, to the total number density N, which is governed by pressure and temperature. The unit of E/N is usually given in Townsend (Td). With either increasing E/N or decreasing O$_2$ fraction, the fraction of high-energy electrons increases, signifying increasing electron “temperature”. The increases in the high-energy component of the electron energy distribution result in larger overlap integrals with the key electronic excitation cross sections, illustrated in Figure 2. In this example, the 10 Td distribution provides power-efficient O$_2$(a) excitation, in that very little power is expended on O$_2$ dissociation; however the poor overlap with the ionization cross section results in a very low ionization rate and consequently low electron number density. The 100 Td distribution gives greater overlap with both the O$_2$(a) excitation cross section and the O$_2$ ionization cross section, but at the expense of increased O$_2$ dissociation. The O$_2$(a) excitation rate is given by the product k$_{exc}$[$e^{-}$][O$_2$], where k$_{exc}$ is the rate coefficient for electron-impact excitation of O$_2$(a). Both k$_{exc}$ and [e$^{-}$], and hence the yield of O$_2$(a), can be considerably enhanced through use of larger E/N and/or lower O$_2$ mole fraction to achieve more energetic electron energy distributions. This illustrates the basic conundrum of electrical O$_2$(a) generation: high power utilization efficiency is optimized by “cold” electron energy distributions (e.g. lower E/N), however high O$_2$(a) production rate and therefore high O$_2$(a) yield require more energetic distributions (e.g. higher E/N).

![Figure 2. Illustration of overlap of electron energy distributions with electron impact excitation cross sections for O$_2$(a) excitation, O$_2$ dissociation to form O + O, and O$_2$ ionization to form O$_2^+$. The electron energy distributions are computed for E/N = 10 and 100 Td, in 10% O$_2$/Ar.](image-url)

4. EXPERIMENTS AND RESULTS

We have used electrodeless, coaxial microwave discharges at 2450 MHz for production of O$_2$(a). For the experiments described herein, we used a microwave induced discharge jet (MIDJet) as the source for O$_2$(a). For earlier, low-power, room temperature discharge-flow experiments, we have used an Evenson-type resonant cavity at 40-120 W power,
with flowing O₂/Ar or O₂/He mixtures at pressures of a few torr, and E/N = 50-100 Td. At higher power and high throughput, we have implemented a coaxial, 1 kW MIDJet device at 40-70 torr O₂/He and ~30-40 Td.

The MIDJet discharge plenum is illustrated in Figure 3. A commercial magnetron and waveguide system deliver 1 to 5 kW of 2450 MHz microwave power via a specially designed coaxial connector. The discharge gas mixture is injected into the plenum via a set of tangential jets to give a swirl flow that confines the discharge plasma near the axis. Typical discharge gas conditions for 1 kW operation are 2-20% O₂ in He, pressures of 35-70 torr, and flow rates of ~40 mmole/s and higher. The discharge-excited gas passes through the discharge exit tubing into the flow reactor, where the excited species other than O₂(a) and O are quickly deactivated by collisional and radiative processes. The discharge plenum and flow reactor are cooled with chilled water. The gas flow passes from the axisymmetric discharge plenum through a transition section into a rectangular flow cross section, and expands supersonically through a two-dimensional contoured nozzle and laser cavity designed for Mach 2-3 flow. Reagents I₂ and NO are injected at various locations in the subsonic flow, and detailed optical diagnostic measurements as described above can be made in both the subsonic and supersonic flow sections. High-reflectance mirrors can be placed on the supersonic cavity for laser power extraction. The PSI supersonic testbed is shown in Figure 4.

Figure 3. Block diagram of 1kW MIDJet.

Figure 4. MIDJet discharge system is at the left and the supersonic laser cavity is at the right.

We used ultra-sensitive, calibrated with absolute standards, to measure the key parameters in their systems. These are summarized in Table 1.

With the MIDJet device, we have demonstrated 20% yield of O₂(a) and nearly 70W of singlet oxygen available for laser output and have scaled the small signal gain by greater than a factor of ten over our initial gain measurements. Indeed, the small signal gain (~ 0.03%/cm) measured in our MIDJet supersonic device is also a factor of 400 greater than our initial measurements of gain in a subsonic lower power microwave system. Here we present results of initial beam quality measurements.
Table 1. Diagnostics Used to Characterize the Discharge Laser Devices

<table>
<thead>
<tr>
<th>Species or Parameter</th>
<th>Method</th>
<th>Wavelength</th>
</tr>
</thead>
<tbody>
<tr>
<td>I*, I, T</td>
<td>TDL Absorption/Gain</td>
<td>1315 nm</td>
</tr>
<tr>
<td>O₂(a,b), I*</td>
<td>InGaAs array emission spectrometer</td>
<td>700-1500 nm</td>
</tr>
<tr>
<td>O</td>
<td>NO-O chemiluminescence</td>
<td>580 nm</td>
</tr>
<tr>
<td>I₂ flow rate</td>
<td>LED micro-absorption</td>
<td>488 nm</td>
</tr>
<tr>
<td>Ozone</td>
<td>UV micro-absorption</td>
<td>254 nm</td>
</tr>
<tr>
<td>I₂ (B)</td>
<td>Visible emission spectrometer</td>
<td>500-700 nm</td>
</tr>
</tbody>
</table>

4.1 Beam and Laser Medium Quality Measurements

We designed and integrated the components for the beam quality measurements into the MIDJet EOIL facility. This system used a near-IR, 250x320 pixel (30µm pitch), InGaAs camera as shown in Figure 5.

![Figure 5. Block diagram of the beam quality measurement system that was integrated into the MIDJet EOIL system.](image)

We completed a comprehensive series of measurements of the beam quality including the propagation factor M². The subsonic flow pressure was ~ 60 Torr and is consistent with a high power EOIL system. We measured M² using two different focal length lenses and determined the beam diameter as a function of distance from the lens.²¹ Our MIDJet EOIL system uses a stable resonator with identical 2 m radius of curvature mirrors spaced approximately 6 cm apart. The active optical pathlength was 5 cm for these tests. The M² propagation parameter measurements provide crucial insight into the medium quality. For example, medium non-uniformities such as density gradients will result in degraded output beams. The goal of these studies was to examine this issue directly. In one series of tests we inserted a 3mm diameter iris within the cavity and close to the output mirror. This restricts the stable resonator to a few TEMₘₙ transverse modes.

We observed a stable TEM₀₀ mode for long periods of time (several minutes). Figure 6 shows a 3-D image of the laser output beam and a cut parallel to the flow direction. The mode is circular and is Gaussian in profile as indicated by the fit to the data.
Figure 6. Sample data of MIDJet-driven EOIL laser output beam.

Figure 7 shows a plot of the beam diameter as a function of the distance \( z-z_0 \) from the lens and a fit to the function:

\[
D = D_0 \left[ 1 + \left( \frac{z - z_0}{z_r} \right)^2 \right]^{1/2}
\]

where \( D \) is the beam diameter at position \( z \),
\( D_0 \) is the beam diameter at its waist, and
\( z_r \) is the Rayleigh range given by \( 4\pi D_0^2/\lambda M^2 \).

Figure 7. Measured diameter of output beam as a function of distance from a focusing lens. A fit to Eq. (1) determines the \( M^2 \) value.
The fit to the data provided an $M^2 = 1.08 \pm 0.01$. This verified that the supersonic laser medium was extremely stable with respect to phase changes/optical path differences. We note that the cavity mirrors were under vacuum and sealed by an O-ring that also served as the alignment jig. Slow, but small variations in the mirror separation during a run caused by thermal changes in the aluminum flow channel forces the mode to gradually change to TEM$_{1,0}$ and TEM$_{0,1}$. We never observed the combination donut mode. Indeed, we observed a pure mode and it was temporally stable over the course of several minutes.

We also measured the beam profile and $M^2$ without the intracavity iris. This provided a gain area of approximately 1 cm$^2$. In this configuration higher order TEM$_{m,n}$ modes with m,n as high as 17,17 were observed as would be expected from a stable resonator extracting power from a volume much greater than that of the TEM$_{0,0}$ mode. The beam diameter at the focus was $\sim (n,m)^{1/2}$ of that of the TEM$_{0,0}$ mode, and the $M^2$ value increased in agreement with stable resonator theory. The important point is that even under these conditions the mode was stable. The increased $M^2$ is caused by higher order modes lasing, not by any medium issues. These results clearly imply that when the optical gain and gain length of EOIL are scaled to allow incorporation of an unstable resonator to extract power from the full EOIL gain volume, excellent beam quality should be produced.

As part of the power scaling of EOIL we have also incorporated a surface catalyst that produces O$_2$(a) from wall reactions involving discharge produced O atoms. We have discussed the catalyst previously$^{22}$ and describe recent results in Ref. 23. These catalytic modules are inserted directly in the flow of the MIDJet discharge. Figure 8 shows photographs of a module.

![Figure 8. Views of catalyst modules. The left panel shows one of two modules containing 12 Macor substrates. The right panel presents details of the geometry of the substrates. The small holes are used to inject the iodine that combines with discharged oxygen to form the coating.](image)

Initial tests on the MIDJet facility using 12 catalyst plates produced a 60% increase in the measured small signal gain. This is a significant result because the output power of the EOIL system will scale at a greater than linear rate as the gain-gainlength product is scaled.

## 5. SUMMARY

We have presented a discussion of recent results relevant to scaling of EOIL to a high power laser system. Using the knowledge gained in this system through a systematic series of studies, we have designed more advanced laser testbeds and demonstrated excellent beam quality and temporally stable output in a supersonic EOIL. The $M^2$ measurements indicate that the medium quality of the EOIL system is sufficient to produce output beams that are nearly diffraction limited. We have also demonstrated significant small signal gain enhancement using the catalytic production of O$_2$(a) in the MIDJet-powered supersonic device. These results, combined with those of References 23 and 24 significantly increase the probability for scaling EOIL to a high power laser.
6. ACKNOWLEDGEMENTS

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REFERENCES


