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1. REPORT DATE (DD-MM-YYYY) 01-08-2006		2. REPORT TYPE Final Report		3. DATES COVERED (From – To) 1 May 2005 - 01-May-06	
4. TITLE AND SUBTITLE Improving COIL Efficiency By Iodine Pre-Dissociation Via Corona Discharge In The Transonic Section Of The Secondary Flow			5a. CONTRACT NUMBER FA8655-05-1-3041		
			5b. GRANT NUMBER		
			5c. PROGRAM ELEMENT NUMBER		
6. AUTHOR(S) Professor Salman Rosenwaks and Dr. Boris Barmashenko			5d. PROJECT NUMBER		
			5d. TASK NUMBER		
			5e. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) Ben-Gurion University of the Negev Ben-Gurion Avenue PO Box 653 Beer-Sheva 84 105 Israel				8. PERFORMING ORGANIZATION REPORT NUMBER N/A	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) EOARD PSC 821 BOX 14 FPO 09421-0014				10. SPONSOR/MONITOR'S ACRONYM(S)	
				11. SPONSOR/MONITOR'S REPORT NUMBER(S) Grant 05-3041	
12. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.					
13. SUPPLEMENTARY NOTES					
14. ABSTRACT This report results from a contract tasking Ben-Gurion University of the Negev as follows: We intend to carry out a comprehensive experimental study of I2 pre-dissociation, based on applying corona discharge in the transonic section of the secondary flow in the COIL supersonic nozzle. The main issues to be addressed are the following: 1. Determination of conditions for maximum dissociation fraction F of I2 caused by the corona discharge. 2. Experimental measurements of the lasing power and small signal gain with and without the iodine pre-dissociation caused by the corona discharge and determination of the power enhancement factor. There will be two stages in the proposed experimental study of I2 pre-dissociation. Facilities: The equipment available at BGU includes a supersonic COIL setup with different mixing nozzles and three diode laser based diagnostic systems for H2O, O2 and I atoms for probing the COIL active medium. Also available are a variety of optical, electronic and computing equipment, including He-Ne lasers, lock-in amplifiers, optical fibers, power supplies, optical windows, halogen lamps and an on-line laboratory computer. Schedule: Report will be delivered at the end of the contract.					
15. SUBJECT TERMS EOARD, Chemical oxygen iodine lasers, Laser physics					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT UL	18. NUMBER OF PAGES 18	19a. NAME OF RESPONSIBLE PERSON DONALD J SMITH
a. REPORT UNCLAS	b. ABSTRACT UNCLAS	c. THIS PAGE UNCLAS			19b. TELEPHONE NUMBER (Include area code) +44 (0)20 7514 4953

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Improving COIL efficiency by iodine pre-dissociation
via corona discharge in the transonic section of the
secondary flow

Contract No. FA8655-0 5-1-30 41

BGU Internal Contract No. 82 311101

Final Report for the Period

1.05.2004 - 30.04.2005

Principal Investigators: Dr. Boris Barmashenko and Prof. Zamik Rosenwaks

I. Abstract

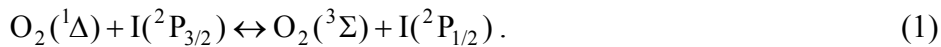
The loss of $O_2(^1\Delta)$, consumed in I_2 dissociation in chemical oxygen iodine laser (COIL) systems, is one of the major obstacles for increasing the chemical efficiency and the power in a given system. We report on a new method for pre-dissociation of I_2 , based on applying corona/glow electrical discharge in the transonic section of the secondary flow in the COIL supersonic nozzle, thus reducing the loss of $O_2(^1\Delta)$. 1.7% of I_2 is dissociated by the discharge resulting in 70% power enhancement at rather high I_2/O_2 ratio, 1.6%, close to the optimal value ($\sim 2.5\%$) for operation of COILs with supersonic mixing.

The following projects have been carried out during the reported period:

1. Design and manufacturing of the iodine mixing nozzle with electrical discharge.
2. Development of a 3D CFD model for flow in injectors with discharge pins.
3. Measurements of I atom production by the discharge.
4. Influence of the discharge on the small signal gain.
5. Influence of the discharge on the laser power.

II. Brief scientific background and objectives of the project

The chemical oxygen-iodine laser (COIL)¹ is the shortest wavelength (1315nm) and the most efficient high-power chemical laser operating to date. Iodine atoms are pumped by a near resonant energy transfer from oxygen molecules in the excited singlet-delta state, $O_2(a^1\Delta_g)$,



Mixing of $O_2(^1\Delta)$ produced in a special chemical generator with I_2 molecules results in the dissociation of molecular iodine to atomic iodine which is subsequently excited via reaction (1). $O_2(^1\Delta)$ is the energy carrier for the COIL, hence the loss of excited $O_2(^1\Delta)$ molecules in the iodine mixing-dissociation zone results in a decrease of laser power.

To get rid of the dissociation losses, Japanese researchers² suggested employing pre-dissociation of I_2 before its injection into the primary flow of singlet oxygen. The pre-dissociation was carried out using microwave discharge in the subsonic section of the secondary I_2/He flow where large pressure resulted in fast recombination of I and as a result the iodine dissociation fraction was $< 50\%$ even at small iodine flow rates ($I_2/O_2 <$

0.8%). For larger I_2/O_2 ratio, close to the optimal 2.5% value (corresponding to the maximum lasing power without pre-dissociation) no iodine dissociation was found.

To preclude recombination of iodine atoms we proposed, following the suggestion of Professor Michael Heaven of Emory University, to pre-dissociate I_2 by corona discharge in the transonic section of the secondary flow, i.e., near the iodine injection holes. Low pressure of the gas, expanding supersonically immediately after the dissociation by the discharge, should result in much slower recombination of I atoms and larger values of the iodine dissociation fraction in the laser section.

Corona discharge was used before (in non-laser applications) to generate radicals and ions in supersonically expanding flows, near the critical cross section, for spectroscopy of jet-cooled ions and radicals³. Using this corona excited supersonic expansion (CESE) technique, a very efficient generation of the radicals was observed with only a few percent of precursor in an inert carrier gas and high pressure of several hundreds Torr. In the COIL the I_2 fraction in the N_2 secondary flow is also only a few percents, and the stagnation pressure of the secondary flow is about 100 Torr, which means that this method can be applied also for iodine pre-dissociation in the COIL.

III. Description of the results

1. Design and manufacturing of the iodine mixing nozzle with discharge.

Our design of the iodine mixing nozzle with discharge was based on the CESE design³. Due to safety reasons (working with high voltage) the iodine-oxygen mixing system was manufactured from ceramic. This nozzle was used for pre-dissociation of I_2 , based on applying discharge in the transonic section of the secondary flow in the COIL supersonic nozzle. Schematic of the nozzle with discharge is shown in Fig. 1.

A non-profiled nozzle with 5 mm throat height and 7.5 mm flow height at the iodine injection location is used for studying the discharge. The nozzle includes one row of 25, 1.4-mm diameter holes in each wall for iodine injection at a 45° angle to the primary flow. Fifty brass pins connected to a high voltage common wire are installed either inside the injection holes of the nozzle or just upstream of the hole inlet. A 2 kV and 1 Amp, DC power supply feeds the discharge. The corona develops in the injection hole and is continued by a glow discharge expanding to the negative electrodes installed in the floor and ceiling of the supersonic section of the flow. To preclude electrical sparks

each of the brass pins was connected to positive voltage in line with $1\text{k}\Omega$ resistor, restricting the current.

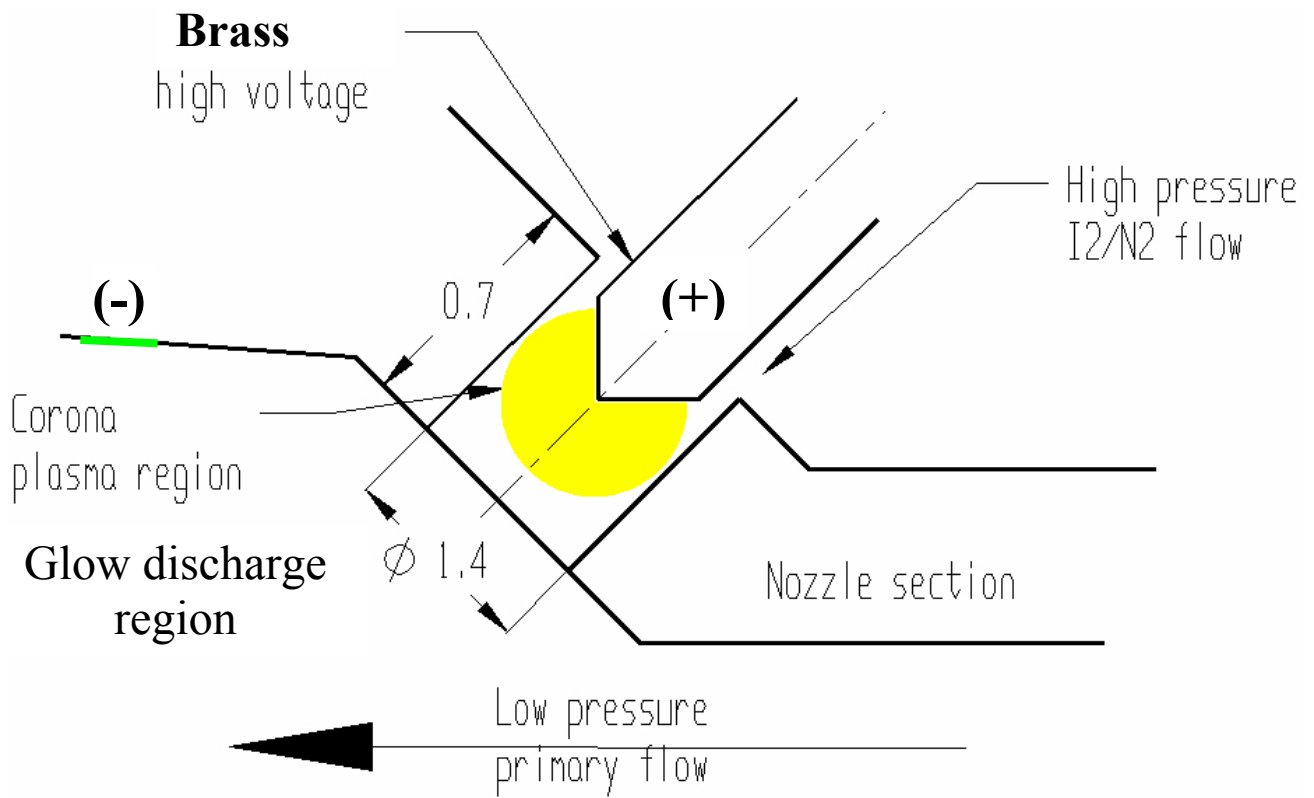


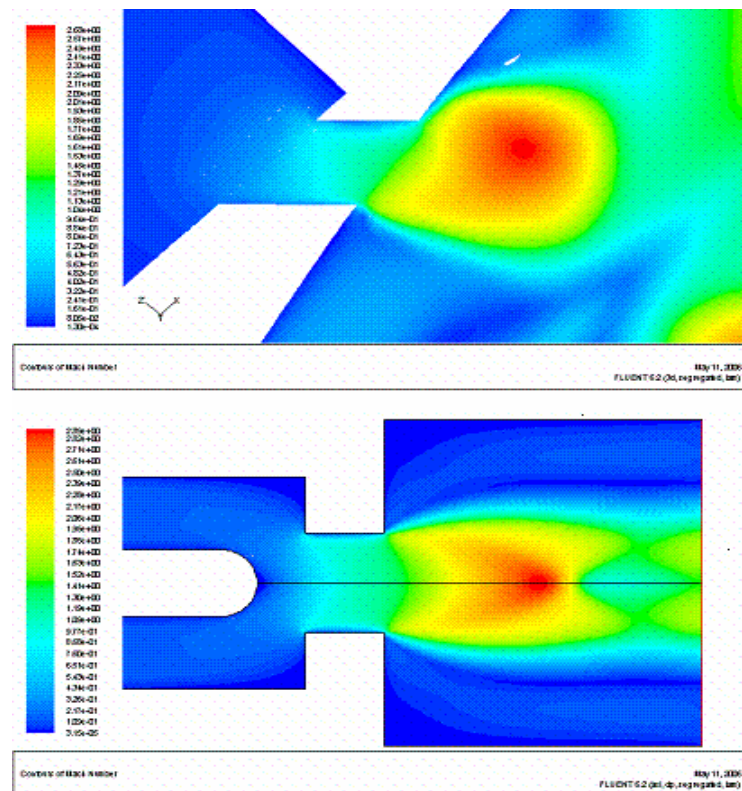
Fig. 1. Schematics of the mixing nozzle with discharge. The spherical yellow region near the tip of the brass electrode (pin) indicates the corona plasma region. Negative brass electrode indicated by green is installed in the floor and ceiling of the supersonic section of the flow.

2. 3D CFD modelling of the flow in the iodine injection hole for different positions of the positive electrode

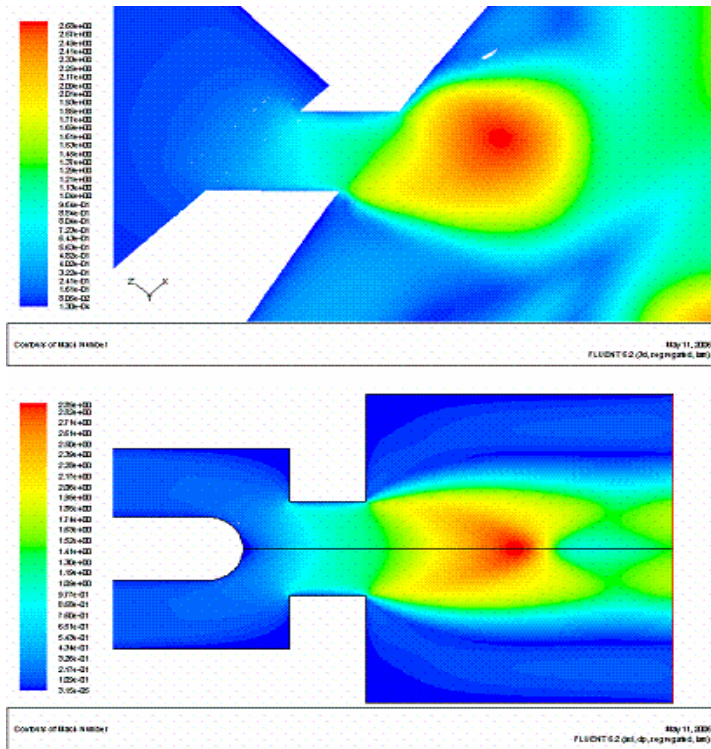
To minimize disturbance of the secondary flow by the brass pin it was necessary to find optimal position of the pin relatively to the injection hole. To do this we used a 3D CFD model of the flow based on the commercial Fluent code. Two different locations of the pin leading edge were tested: 0.6 mm upstream and 0.5 mm downstream of the injector inlet. For the first case spatial distributions of the pressure p and Mach number M as compared with the case when the pin is absent are shown in Fig. 2 a and b,

respectively. It is seen that the flow field is very similar to the case when the pin is absent and the pin does not block the secondary flow. For the second position of the pin the spatial distributions of p and M are shown in Fig. 3 a and b, respectively. Unlike the first case here the pin blocks the secondary flow, strongly decreasing the critical cross section which is located in the space between the pin outer and hole inner surfaces.

Hence the first location of the pin upstream of the injector inlet is better for the flow dynamics since it provides for the same jet parameters at the injector exit as the flow without the pin. For the second pin location the jet penetration into the primary flow is too deep resulting in large primary flow stagnation pressure and singlet oxygen losses.

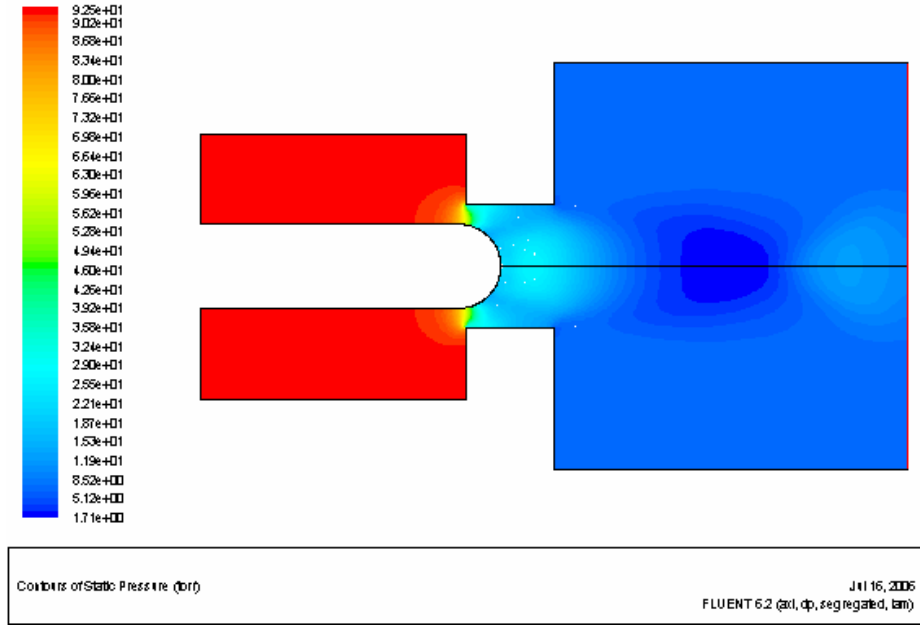


a

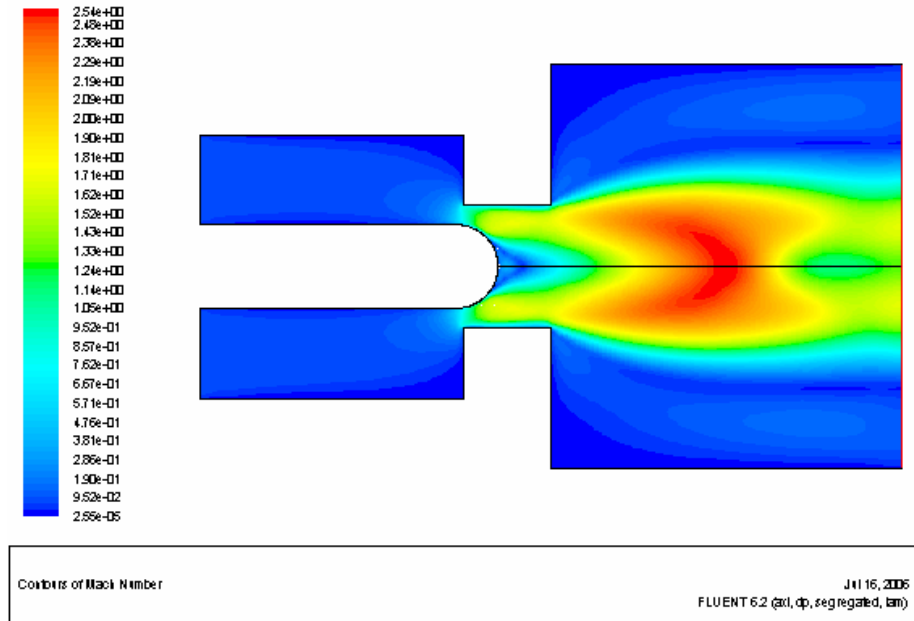


b

Fig. 2. Spatial distributions of the static pressure (a) and Mach number (b) for the brass pin leading edge located 0.6 mm upstream of the injection hole inlet as compared with the flow without pin (upper panels in both (a) and (b)). **Note that the upper panels were calculated for non-symmetric flow and the lower for symmetric).**



a



b

Fig. 3. Spatial distributions of the static pressure (a) and Mach number (b) for the brass pin leading edge located 0.5 mm downstream of the injection hole.

3. Measurements of I atom production by electrical discharge.

In the first stage, conditions were determined for maximum I atoms production by the discharge. In these "cold" experiments $O_2(^1\Delta)$ in the primary flow was replaced by N_2 to preclude the process of I_2 dissociation by $O_2(^1\Delta)$. The positive electrode leading edge

was located 0.5 mm upstream from the injector inlet. As shown above (section 2) this geometry provides for the best flow conditions for the laser operation. A glow discharge of violet-green color was seen in the supersonic section of the flow. Sparks were absent but the discharge was non-steady and non-uniform both along and across the flow. V-I characteristic of the discharge for typical range of the currents (0.5-1 A) is shown in Fig. 4.

I atoms were probed using 1315 nm beam from the diode laser based diagnostic system developed by Physical Sciences Incorporation. The system monitors the absorption coefficient k for the $I^*(5p^5 \ ^2P_{1/2}, F=3) \rightarrow I(5p^5 \ ^2P_{3/2}, F=4)$ transition at 1315 nm. The laser frequency is scanned over the I transition, monitoring the absorption profile. The temperature T of the gas in the cavity is found from the Doppler linewidth $\Delta\nu_D$, the later being determined by fitting the Voigt function to the experimental gain. Typical absorption line is shown in Fig. 5. It is seen that the value of the absorption coefficient is smaller than 0.05%/cm, this is due to small dissociation fraction of I_2 caused by the discharge.

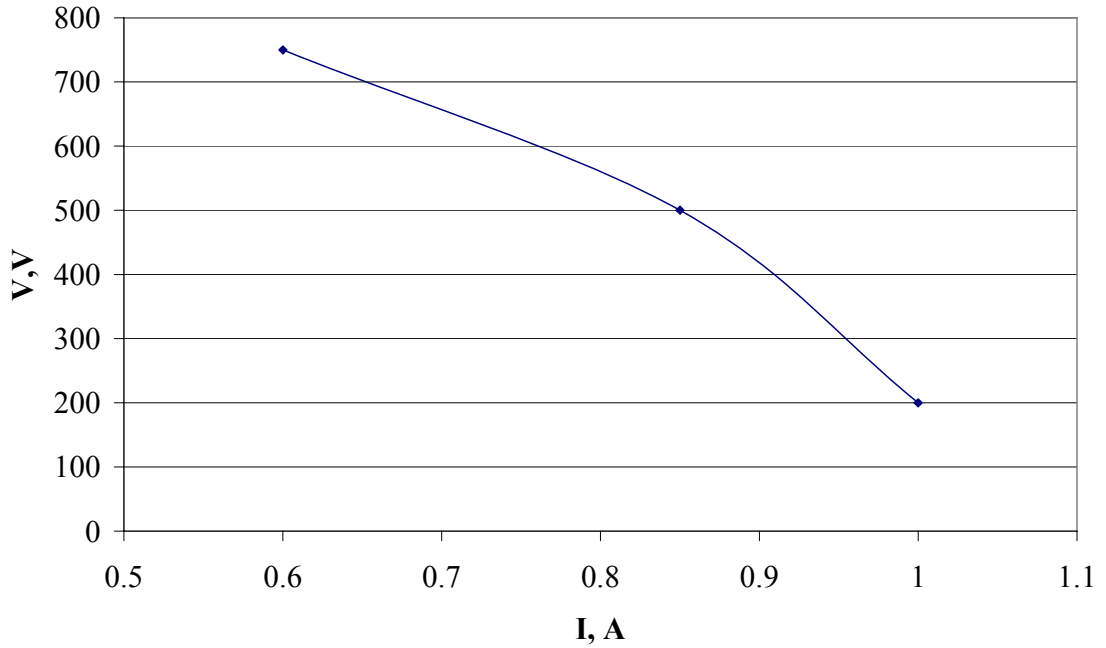


Fig. 4. V-I characteristic of the discharge for the brass pin located 0.5 mm upstream of the injector inlet, $(nN_2)_p = 21$ mmole/s, $(nN_2)_s = 28$ mmole/s and $nI_2 = 0.4$ mmole/s.

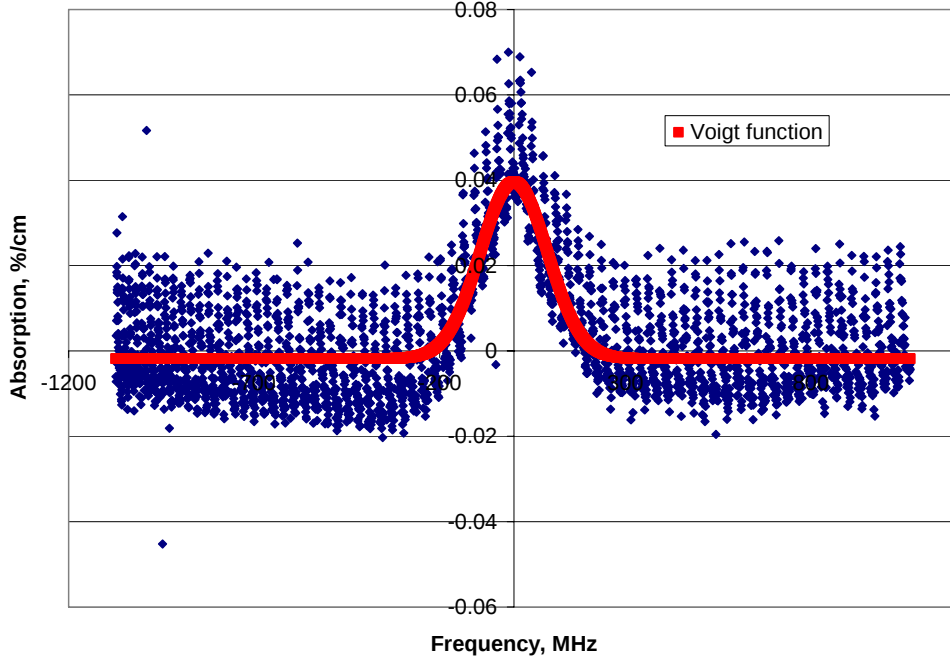


Fig. 5. Absorption line of I atoms produced by the discharge.

$[I]$ is determined from measured k and T assuming equilibrium between I^* and $O_2(^1\Delta)$ in the pumping reaction (3):

$$k_0 = \frac{7}{12} \sigma_0 \left(\frac{300}{T} \right)^{1/2} \frac{[I]}{2}, \quad (2)$$

where $\sigma_0 = 1.29 \times 10^{-17} \text{ cm}^2$ and k_0 are the stimulated emission cross section for the I atom transition and absorption coefficient at the line center, respectively. The dissociation fraction of I_2 molecules can be estimated as:

$$F = \frac{[I]}{2(p/kT)(\chi_{I_2})_{av}}, \quad (3)$$

where $(\chi_{I_2})_{av} \equiv (n_{I_2})/n$ is the average molar fraction calculated from the measured flow rate under assumption of homogeneous I_2 distribution across the flow and n is the total flow rate.

The flow parameters and highest values of $[I]$ and F for the nozzles applied in our experiments are presented in Table I. For optimal flow rates corresponding to the maximum lasing power and chemical efficiency of the COIL (run 1) the values of $[I]$ and F are rather low, in particular the value of F is smaller than 2%. For smaller primary

and secondary flow rates corresponding to smaller pressures (run 2) larger values of $[I]$ and F are obtained, however, these flow rates are not optimal for the COIL operation.

Table 1. Maximum values of $[I]$ and F for different flow parameters.

Run	$(nN_2)_p$, mmole/s	$(nN_2)_s$, mmole/s	nI_2 , mmole/s	Voltage, V	Current, A	$[I]$, cm^{-3}	F , %
1	21	28	0.4	200	1	4×10^{13}	1.7
2	10	13	0.3	50	1	9×10^{13}	4

Spatial dependence of $[I]$ across the flow for runs 1 and 2 is shown in Fig. 6. It is seen that these dependence is not uniform and more I atoms are produced above the optical axis which is due to slightly non-symmetrical geometry of the electrodes in the upper and lower nozzle walls caused by both non-perfect manufacturing and non-uniformity of the discharge.

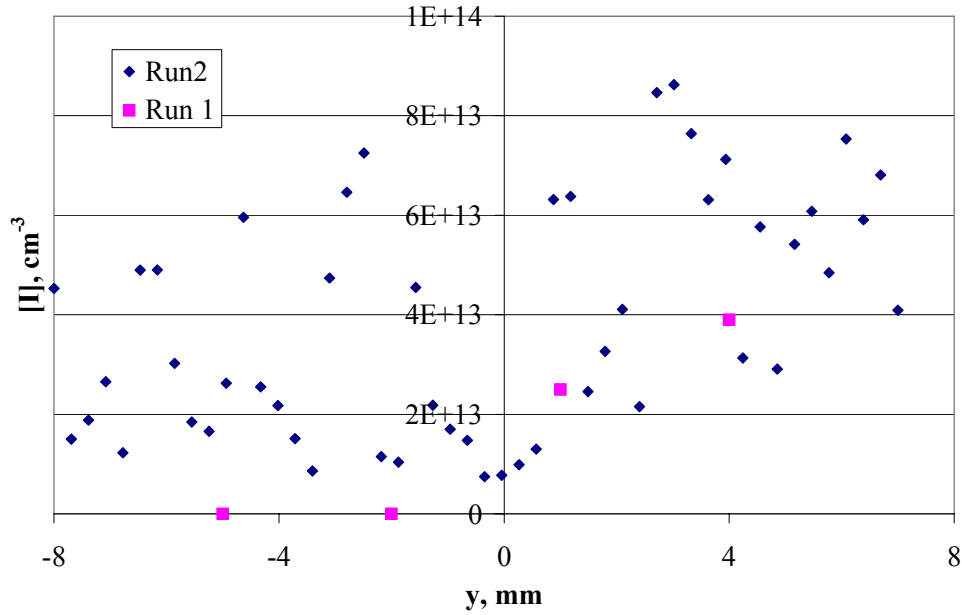


Fig. 6. Spatial distribution of $[I]$ produced by discharge across the flow at the optical resonator for runs 1 and 2.

As shown in Fig. 7, the maximum values of $[I]$ and F increase with increasing the discharge current I . Unfortunately the maximum current of our power supply is 1 A and it is impossible to increase the current and to get larger values of iodine dissociation fraction

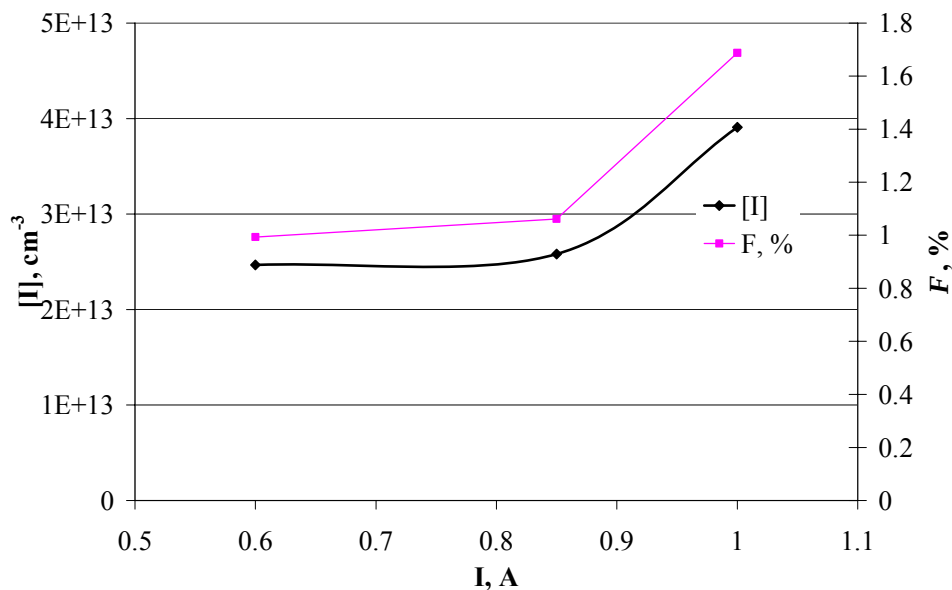


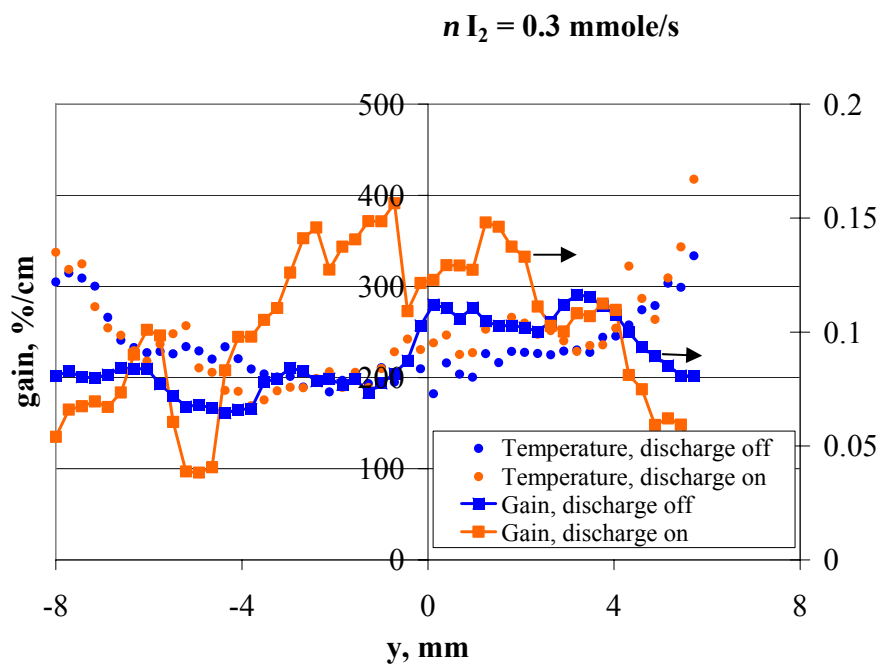
Fig. 7. Dependence of maximum $[I]$ and F on the discharge current I for the same flow conditions as in run 1.

4. Influence of the discharge on the small signal gain.

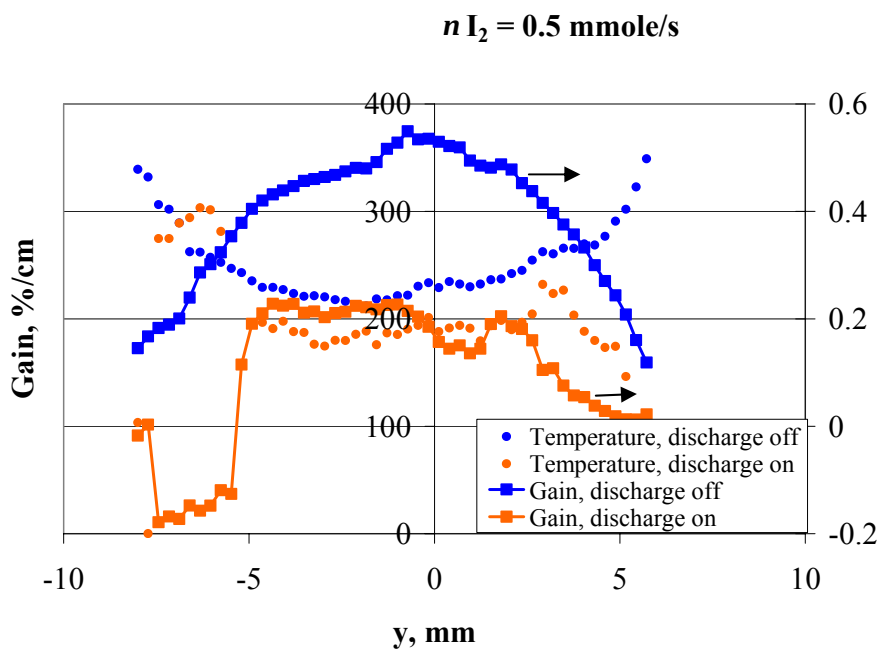
The influence of the discharge on the gain was studied in hot runs where $\text{O}_2(^1\Delta)$ produced in the chemical generator was the primary gas. The gain and temperature distributions across the flow are shown in Fig. 7 a and b for two different $n\text{I}_2$. It is seen that for small $n\text{I}_2$ switching the discharge on results in increasing the gain, whereas for large $n\text{I}_2$ the gain decreases as the discharge is switched on, the temperatures with and without discharge being almost the same.

Fig. 8 shows dependence of the maximum gain (over the flow cross section) on $n\text{I}_2$ with the discharge on and off. It is seen that application of the discharge results in gain enhancement for $n\text{I}_2 < 0.4$ mmole/s, whereas for larger $n\text{I}_2$ the gain decreases as the discharge is applied. The gain enhancement observed for small $n\text{I}_2$ can be explained by Heidner's mechanism of I_2 dissociation. For this mechanism iodine dissociation is an autocatalytic chain reaction accelerated by I^* formed in reaction (1). Hence the dissociation rate is proportional to $[I]$ and for small $n\text{I}_2$ formation of I atoms caused by electrical discharge results in much faster dissociation and larger values of the gain than in the absence of the discharge. For higher $n\text{I}_2$ the dissociation enhanced by the discharge is completed upstream of the optical axis (where the gain is measured). Quenching of I atoms results in smaller values of the gain than for the case of slower dissociation without discharge when the gain at the optical axis continues to increase

with increasing nI_2 . Another reason for large I quenching and smaller gain with applied discharge is the fact that the static pressure in the resonator higher than that without discharge (see Fig. 8). The pressure increase may be due to heat release in the flow, caused by the discharge.



a



b

Fig. 7 The gain and temperature distributions across the flow in the hot runs for the discharge on and off; the chlorine and secondary nitrogen rates are 17 and 28 mmole/s, respectively, nI_2 is 0.3 and 0.5 mmole/s for figures (a) and (b), respectively.

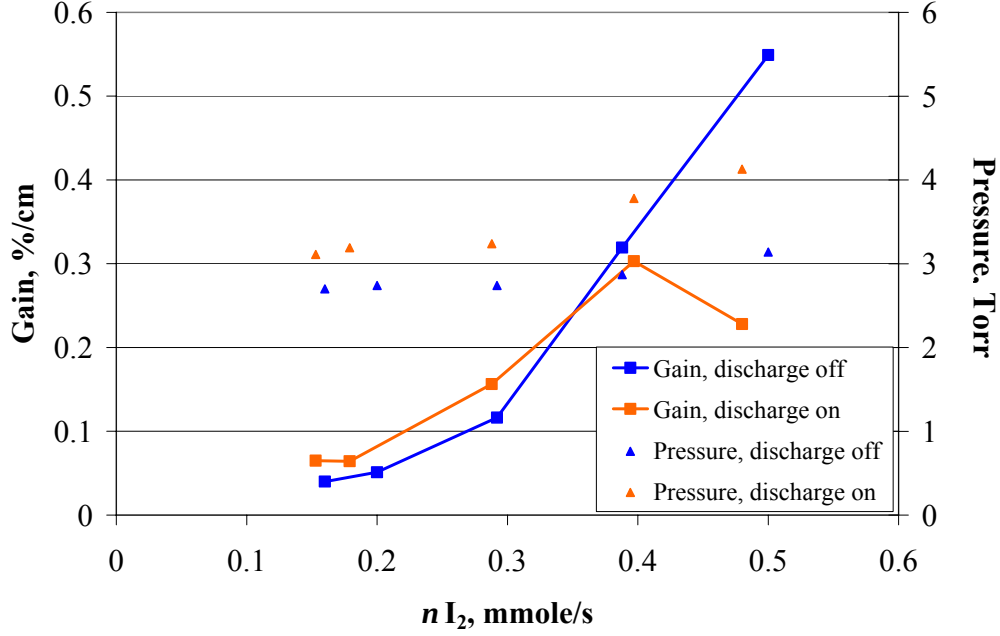


Fig. 8. Maximum gain (over the flow cross section) and the pressure with and without discharge as a function of the iodine flow rate.

5. Influence of the discharge on the laser power.

Fig. 9 shows the temporal evolution of the lasing power for the discharge switching for different nI_2 . Just as the gain, the power is increased by the discharge up to $nI_2 = 0.4$ mmole/s and decreases for larger nI_2 . Dependence of the power with and without discharge on nI_2 is shown in Fig. 10. The same figure shows the ratio of the powers with discharge on and off. It is seen that the maximum power enhancement reaches $\sim 70\%$ at small $nI_2 = 0.24$ mmole/s. For larger nI_2 the power enhancement decreases and is negative for $nI_2 \geq 0.4$ mmole/s.

70% power enhancement at $I_2/O_2 = 1.6\%$ obtained using DC corona/glow discharge is larger than the 50% enhancement obtained in Ref. 2 at very small $I_2/O_2 = 0.8\%$ using microwave discharge applied in the subsonic section of the secondary flow. Hence the present discharge scheme has an advantage over the scheme used in Ref. 2.

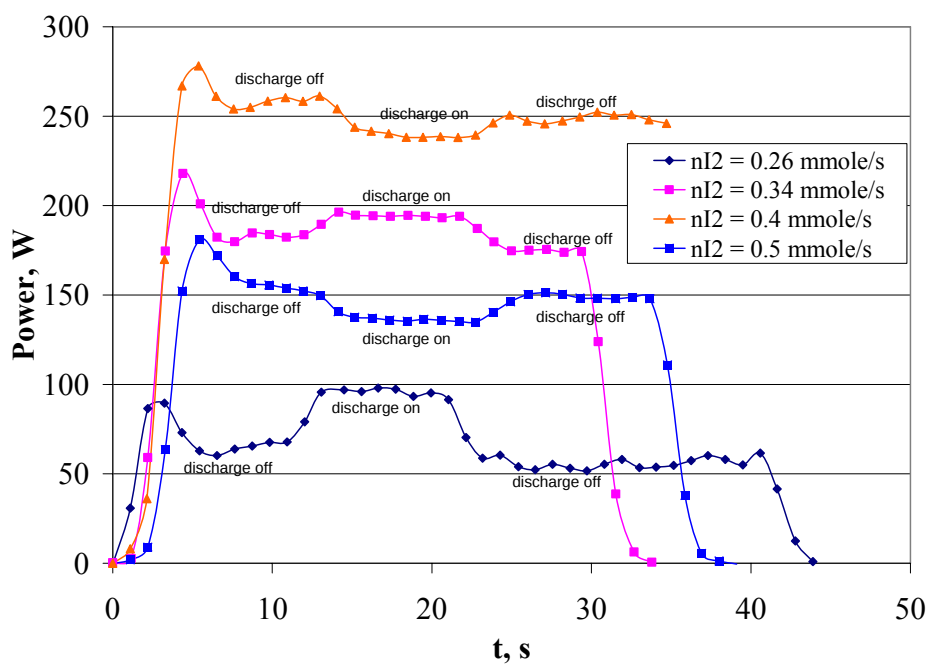


Fig. 9. Temporal behavior of the power with discharge on and off for different iodine flow rates

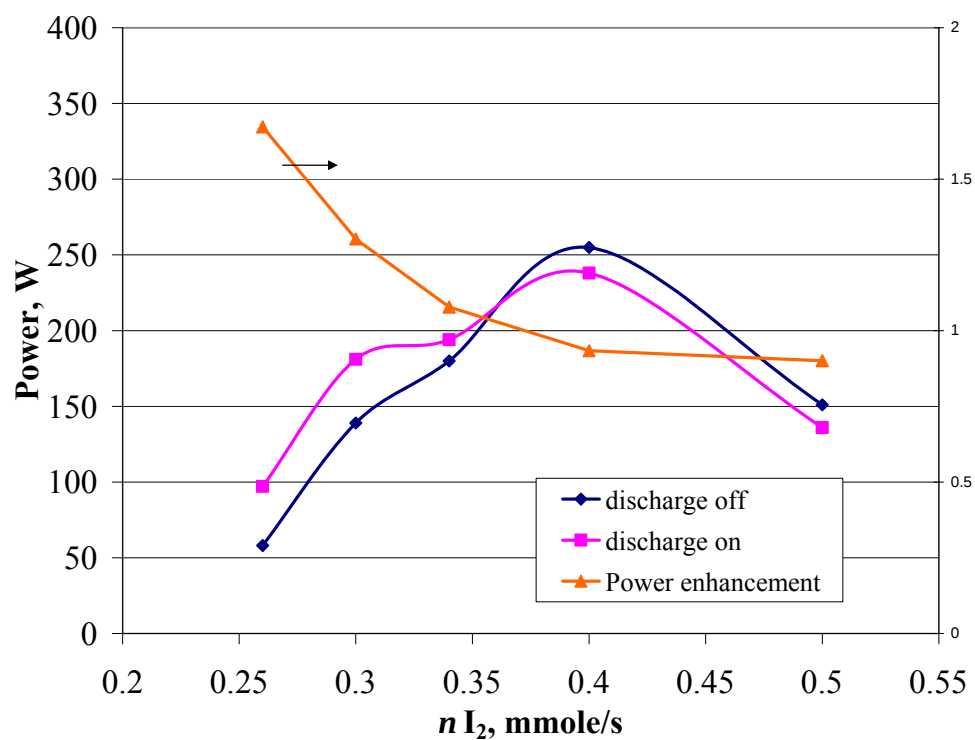


Fig. 10. Dependence of the power and the power enhancement on nI_2 with the discharge on and off.

IV. Summary and recommendations

A detailed study of the COIL with iodine pre-dissociation caused by the DC discharge applied in the transonic and supersonic sections of the secondary flow was carried out. Up to several percents of I_2 can be dissociated by the discharge; however, for the optimal primary and secondary flow rates the dissociation fraction of I_2 caused by the discharge is $\sim 1.5 - 2\%$.

Application of the discharge with relatively high electrical power (200 V and 1 A) results in gain and laser power enhancement for small $nI_2 < 0.4$, whereas for larger nI_2 the gain and power decrease as the discharge is applied. 70% power enhancement by the discharge is obtained at rather high $I_2/O_2 = 1.6\%$, close to the optimal value ($\sim 2.5\%$) for COILs with supersonic mixing, though the maximum power (achieved for large iodine flow rates) without discharge is still larger than that with discharge.

To increase the I_2 dissociation fraction and power enhancement it is necessary to optimize the electrical power of the discharge, secondary flow rate and the distance between the injection location and the resonator optical axis. Experiments toward achieving these goals are underway in our lab.

¹W. E. McDermott, N. R. Pchelkin, D. J. Benard, R. R. Bousek, Appl. Phys. Lett., 32, 469 (1978).

²H. Okamoto, T. Hirata, K. Shinoda, S. Takeda, D. Sugimoto and M. Endo, AIAA Paper 2000-4032.

³P. C. Engelking, Rev. Sci. Instrum. 57, 2274(1986).

V. Supplements

1. List of the people participating in the project

The list includes only the people participating in the research (it does not include the workers of different workshops that helped in building and maintenance of the experimental setup):

Prof. Zamik Rosenwaks, principal investigator

Dr. Boris Barmashenko, principal investigator

Dr. Karol Waichman, investigator

Mr. Victor Rybalkin, investigator (Ph. D. student)

Mr. Arje Katz, investigator (Ph. D. student)

Mr. Zadok Dahan (M. Sc. student)

2. List of the technical documents during the reported period

The technical documents that appeared during the reported period include one report submitted to the EOARD 6 months after the onset of the research and the papers published or during the reported period:

1. V. Rybalkin, A. Katz, B. D. Barmashenko, and S. Rosenwaks, "Parametric study of a highly efficient chemical oxygen-iodine laser with supersonic mixing of iodine and oxygen ," *J. Appl. Phys.*, **80**, 023106 (2005).
2. S. Rosenwaks, V. Rybalkin, A. Katz and B. D. Barmashenko, "Diagnostic Studies of Ben-Gurion University High Efficiency Supersonic COIL," *AIAA* 2005-5165 (2005)
3. B. D. Barmashenko, V. Rybalkin, A. Katz and S. Rosenwaks, "Detailed gain measurements and analysis of a highly efficient supersonic COIL," in International Conference on Lasers, Applications, and Technologies 2005: High-Power Lasers and Applications, Proc. SPIE, 2004, vol. 6053, 605306 (2006).
4. V. Rybalkin, A. Katz, K. Waichman, D. Vingurt, Z. Dahan, B.D. Barmashenko and S. Rosenwaks, "How many $O_2(^1\Delta)$ molecules are consumed per dissociated I_2 in chemical oxygen-iodine lasers?", *Appl. Phys. Lett.*, vol. 89, 021115 (2006).

VI. Acknowledgment

This material is based upon work supported by the EOARD under Contract No. FA8655-0 5-1-30 41.