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A contribution of COIL Laboratory in Prague to the Chemical Oxygen-Iodine Laser research and development

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ABSTRACT

The key results gathered in the COIL Laboratory of the Institute of Physics AS in the Czech Republic since 1985 to date on the experimental and theoretical investigation of Chemical Oxygen-Iodine Laser (COIL), and related problems are reviewed in a certain context of historical perspective of the COIL research and development.

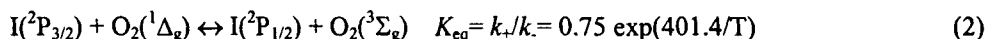
Keywords: chemical lasers, oxygen-iodine laser, singlet oxygen, atomic iodine

1. INTRODUCTION

Laser emission at 1.315 μm from magnetic-dipole transition between spin-orbit levels of the ground state configuration of iodine atom



was first observed during flash photolysis of alkyl iodides, and since its discovery by Kasper and Pimentel¹ in 1964, the iodine photodissociation laser has been developed into systems capable of very high energies. The Prague Asterix Laser System (PALS) is currently one of the largest such facilities in the world, delivering on the target a focus power density up to 10^{16} W/cm^2 in 400 ps (at the fundamental wavelength).² Lasing mechanism of the Chemical Oxygen-Iodine Laser (COIL) follows the same physical principle, only the $^2P_{1/2}$ upper level is pumped via the energy of singlet delta state of molecular oxygen, $\text{O}_2(^1\Delta_g)$. Unique physical and chemical properties of this metastable species³ that is able to transfer its energy to iodine atom in a near-resonant process, and can be generated chemically with very high efficiency have brought about the exceptional position of COIL among other chemical lasers. The pumping mechanism discovered by Derwent *et al.*⁴ in the seventies, is represented by the reverse reaction process with temperature dependent equilibrium constant



Twenty-five years have passed in 2002 since McDermott and his colleagues⁵ from the Air Force Weapons Laboratory (the Air Force Research Laboratory nowadays) demonstrated the first COIL generation on the milliwatt level. A multihundred-kilowatt version of the COIL facility has been developed and tested for the Airborne Laser program⁶ since that time. In the meantime, a tremendous theoretical and experimental work has been devoted to understand comprehensively this laser system to bring it to the present technological level. The Prague laboratory joined this investigation in 1985. Theoretical and experimental efforts have been focused on partial problems in the COIL operation that have concerned, for example, the $\text{O}_2(^1\Delta_g)$ generation for both subsonic and supersonic COIL, the chemical generation of atomic iodine for a COIL, laser performance characteristics in CW and pulsed regime, etc. A brief cross-section through results of this investigation is presented below.

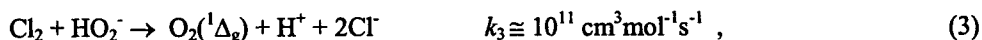
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2. SINGLET OXYGEN GENERATION

2.1. Mechanism of $O_2(^1\Delta_g)$ loss in a low-pressure chemical generator

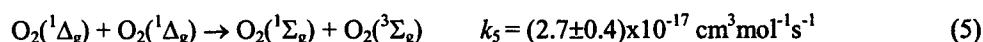
The 25th commemorative COIL session permits mentioning the interesting result following from our "pioneer" work done on the $O_2(^1\Delta_g)$ generation by the reaction⁷



and the mechanism of $O_2(^1\Delta)$ loss in a low-pressure bubbler-type (sparger) generators⁸⁻¹⁰ employed for driving the COIL in subsonic flow regime. The $O_2(^1\Delta_g)$ formed in the liquid film surrounding the bubbled chlorine through the BHP has to diffuse into the gas bubble during the time that the bubble is moving up through the head of liquid (basic solution of hydrogen peroxide, BHP). The gas pressure (consisting of reacting chlorine and formed singlet oxygen) within the two-phase (g-l) bubbling mixture in the reactor column is substantially increased due to a hydrostatic pressure of the liquid column than in the gas space above the liquid head. For example, if the O_2 pressure in the generator plenum is 1 Torr, then the 8 cm liquid head provides the hydrostatic pressure of 8 Torr on gas bubbles at the reactor bottom. By this effect, the loss of $O_2(^1\Delta_g)$ in gas bubbles at the bottom is 64 times higher. A mathematical model for estimation of these loss-processes in sparger-type generators was developed considering a variation in the volumetric fraction of gas in g-l mixture with a distance from reactor bottom, as well as the effect of water vapour pressure in gas bubbles on the $O_2(^1\Delta_g)$ loss.⁹ This simplified model is based on the mass balance for $O_2(^1\Delta_g)$ over a differential layer dy of the g-l mixture following from the relation

$$-N_A n d\eta = r_4 \beta S dy \quad (4)$$

N_A - the Avogadro constant, n - molar flow rate of chlorine or oxygen, η - $O_2(^1\Delta_g)$ yield from layer dy (y - vertical coordinate specifying the distance from reactor bottom), S - reactor cross section, r_4 - rate constant of reaction (5).¹¹



The β factor is the volumetric fraction of gas in the g-l mixture evaluated from the height of two-phase (foamy) mixture (h) and a liquid height without gas in the reactor (h_0), $\beta = 1 - h_0/h$. There are two main $O_2(^1\Delta_g)$ deactivation processes in these generators. First, the $O_2(^1\Delta_g)$ in the bubble diffuses back to the liquid surrounding it, is attached and deactivated. Secondly, a deactivation/pooling $O_2(^1\Delta_g)$ self-quenching process occurs in the gas phase (bubble) itself by the reaction (5). This process is pressure dependent (quadratic in $O_2(^1\Delta_g)$ concentration), and consequently strongly accelerated within g-l mixture due to the hydrostatic pressure of liquid (in the above given example, by two orders of magnitude). Contrariwise, if the bubble residence time in BHP is too short, not all Cl_2 is converted into $O_2(^1\Delta_g)$ and generator yield is also low. The results of this research are remembered in Fig. 1.

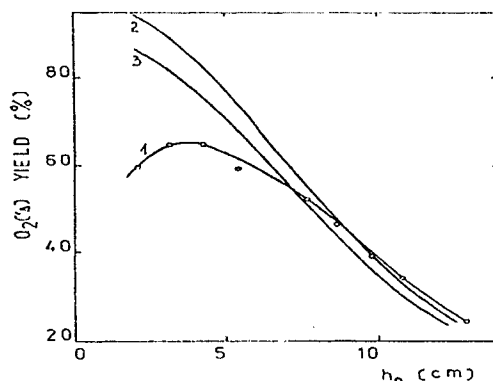


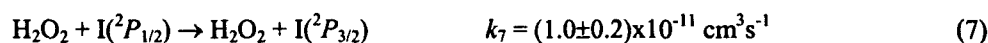
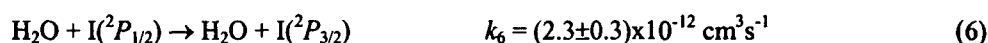
Fig. 1. $O_2(^1\Delta_g)$ yield as a function of BHP height in reactor. 1 - experimental curve, 2 - calculated curve including $O_2(^1\Delta)$ loss by reaction (5), 3 - calculated curve including another $O_2(^1\Delta)$ losses (in liquid phase, quenching on reactor wall, transport in duct)

It is evident that this type of singlet oxygen generator can operate at moderate/low pressure, and only way to achieve high mass flow is to increase the generator surface area resulting in a larger O₂ collection plenum. This finding was afterwards considered in designs of sparer-type generators.

2.2. Generation of O₂(¹Δ_g) in a high-pressure jet SOG

Development of the supersonic COIL during early ninetieth called for a high-pressure O₂(¹Δ_g) generators. We concentrated on the investigation of the jet-type SOG that was set up first in the Samara COIL laboratory.¹² A large surface area of liquid in the form of liquid jets for performing the reaction (3) allowed a substantial reduction of the generator volume (dimensions), and consequently the O₂(¹Δ_g) loss in the gas phase. Velocity of liquid jets is very high (5-20 m/s), which ensures a fast renovation of the gas-liquid boundary, and so minimizes HO₂⁻ ions depletion from the surface layer. It contributes to lowering the O₂(¹Δ_g) loss also in the liquid phase. Our effort in the investigation of this generator was focused on some problems in operation of this generator.¹³⁻¹⁶ Experimental results obtained on the small-scale pilot device concerned, for example, the influence of different configurations and diameters of holes (0.3, 0.5, and 0.8 mm) in the BHP injector on the jets stability, and a detailed investigation of water vapour estimation in gas exiting the generator. We searched for conditions, at which the water vapour content was minimized.

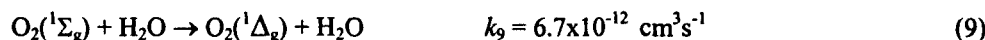
Water formed during O₂(¹Δ_g) generation causes together with peroxide vapor a significant problem in the COIL operation because it quenches the excited state of atomic iodine in very fast processes¹⁷



In a supersonic laser, water can cause also undesirable fluid-dynamic effects due to its condensation during the gas expansion cooling.¹⁸ Water vapor concentration in gas was estimated by the relation (8) derived from the mass balance of O₂(¹Σ_g) state.¹⁹

$$c_{\text{H}_2\text{O}} = k_4 c_{\Delta}^2 / k_9 c_{\Sigma} \quad (8)$$

This species is formed in the pooling reaction (5), and is very efficiently quenched by water molecule.²⁰



Water partial pressure was evaluated from the emissions of O₂(¹Σ_g) and O₂(¹Δ_g) states for different BHP composition, BHP jets temperature, varying Cl₂ flow rate, and different BHP injector configurations. The experimentally obtained data were compared with calculated values of the saturated water vapour pressure above the BHP (for given BHP compositions and temperatures). The both values were nearly the same. The relative water vapour content was well decreasing on increasing the generator pressure. It was the experimental proof that employing the high-pressure jet SOG in the COIL operation reduces a detrimental effect of water.

2.3. The Einstein coefficient for O₂(¹Δ_g) fundamental emission

Work on the development of high-pressure jet-type chemical generator of O₂(¹Δ_g) and the above-described parametric investigations, and simultaneously in the literature opened discussions on the right value of the Einstein coefficient for O₂(¹Δ_g) emission at 1.27 μm (*A*-coefficient) triggered our interest in verification of this coefficient. Mlynczak and Nesbit²¹ challenged the Badger *et al*'s²² value of 2.58 × 10⁻⁴ s⁻¹ when they reported a new value of 1.47 × 10⁻⁴ s⁻¹. The radiative lifetime of O₂(¹Δ_g), τ_Δ^{rad}, ~65 min (a half-lifetime ~45 min) considered so far in the COIL, increased to ~113 min. A detailed insight into this problem revealed that published values of the Einstein *A*-coefficient evaluated by means of various theoretical approaches and experimental techniques varied in a wide range resulting in τ_Δ^{rad} from ~53 to ~151 min.^{23,24} This fact could make the evaluation of O₂(¹Δ_g) concentration in COIL questionable.

To verify a rightness or faultiness of the *A*-value used so far, the way of employing two independent experimental techniques simultaneously was chosen.²⁵ The electron paramagnetic resonance (EPR) spectroscopy was one method, and the optical emission spectroscopy was the second one supplemented with the mathematical model developed for the

evaluation of $O_2(^1\Delta_g)$ concentration from the emission measurements. In this model, we assumed that $O_2(^1\Delta_g)$ molecules are distributed uniformly in the optical cell emitting photons in all directions with a rate, r ,

$$r = A_\Delta c_\Delta^{ES} = A_\Delta P_\Delta / RT \quad (10)$$

A_Δ - Einstein coefficient for spontaneous emission of $O_2(^1\Delta_g)$, P_Δ - $O_2(^1\Delta_g)$ partial pressure.

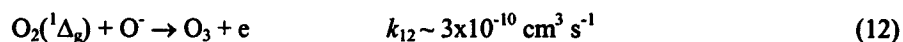
The $O_2(^1\Delta_g)$ concentration, c_Δ^{ES} , was evaluated from the measured signal of the fundamental emission at 1.27 μm and declared sensitivity of the photodiode, and the calculated ratio of the $O_2(^1\Delta_g)$ emission detected by the photodiode and the emission of all $O_2(^1\Delta_g)$ molecules present in the detection cell. The proposed mathematical model was used to evaluate this ratio, in which a photocurrent of the photodiode was directly proportional to a value of the Einstein A -coefficient substituted into relation (10).²⁵ A quantitative evaluation of the $O_2(^1\Delta_g)$ concentration by means of the EPR spectroscopy was based on the "double integral method" of EPR spectra processing by the relation²⁵

$$c_\Delta^{EPR} = k_\Delta \int_{-\infty-\infty}^{\infty} \int_0^B y_\Delta dB dB \quad (11)$$

By comparison of the $O_2(^1\Delta_g)$ concentrations obtained contemporarily by both experimental methods and supplemented by the mathematical model, a new value of the Einstein A -coefficient of $(2.24 \pm 0.4) \times 10^{-4} \text{ s}^{-1}$ was obtained. The corresponding $O_2(^1\Delta_g)$ radiative lifetime is ~ 74 min. Shortly after our first reporting this result, Newman *et al.*²⁶ published the value of $(2.19 \pm 0.07) \times 10^{-4} \text{ s}^{-1}$ giving the $\tau_\Delta^{rad} \sim 76$ min.

2.4. Generation of $O_2(^1\Delta_g)$ in a discharge plasma

The theoreticians of our department performed during the eightieth the fundamental investigation on the physical kinetics of high-frequency (hf) glow discharge in oxygen with respect to generation of the $a^1\Delta_g$ state for pumping the iodine laser.²⁷ Coming out from the properties of the electron gas described by solution of the Boltzmann equation for the electron distribution function, the $O_2(^1\Delta_g)$ concentration in the plasma was calculated with precondition that population of this electronic level is driven by electron collisions only. The upper concentration limit of $O_2(^1\Delta_g)$ achievable in this reaction system was evaluated from the statistical balance between the production and loss processes. The calculated relative population of $a^1\Delta_g$ state, X_Δ/N , is shown in Fig. 2 as a function of the hf field parameters E_{eff}/N and ω (E_{eff} - effective value of hf field, N - concentration of neutral molecules, ω - frequency). It can be seen that the yield of $O_2(^1\Delta_g)$ in hf discharge plasma in pure oxygen can hardly exceed 15% in the chosen range of parameters. Moreover, it is the maximum $O_2(^1\Delta_g)$ concentration that might be still reduced by collisions with negative ions present in the plasma, e.g.²⁷



Outside the region of active discharge, the relative $O_2(^1\Delta_g)$ concentration is even lower (only 3 to 10% in the pressure range of 1-10 Torr). According to this studies, the hf discharge unlikely might produce the $a^1\Delta_g$ state in the amount sufficient to sustain the operation of iodine laser pumped by the energy of such generated singlet oxygen. This conclusion was supported by a subsequently performed analysis by Swift²⁸ of the kinetic processes that accompany the $O_2(^1\Delta_g)$ generation in microwave discharge plasma in pressure range of 1-10 Torr. By these calculations, a maximum attainable relative concentration of $O_2(^1\Delta_g)$ was 10%.

A current experimental research on plasma discharge generation of $O_2(^1\Delta_g)$ for pumping the iodine laser (Discharge Oxygen-Iodine Laser, DOIL) is performed by our colleague J. Schmiedberger in his long-time co-operation with Japanese Institutes. The last results were obtained with the generation of radio-frequency plasma of O_2+N_2+NO mixture in the hollow electrode jet generator.²⁹ The generator is schematically shown in Fig. 3. The plasma jet was extra chilled reactively with transversely injected mixture of $Ar+NO_2$. Adding NO to oxygen enhanced the $O_2(^1\Delta_g)$ production up to 20% through the energy transfer from electronically excited state of NO molecule to O_2 . Another increase was achieved when NO_2 was injected into the reaction system, which caused via certain kinetic processes a lowering in a content of O

and O_3 species in the gas mixture. The $O_2(^1\Delta_g)$ with 32% yield was generated at pressure of 0.43 Torr. In combination of this rf plasma generator with a transversally injected molecular iodine, atomic iodine emission at $1.315 \mu m$ was detected at the laser output mirror by the spectrum analyser. This emission was recalculated to the output power of $\sim 3 \text{ nW}$.²⁹

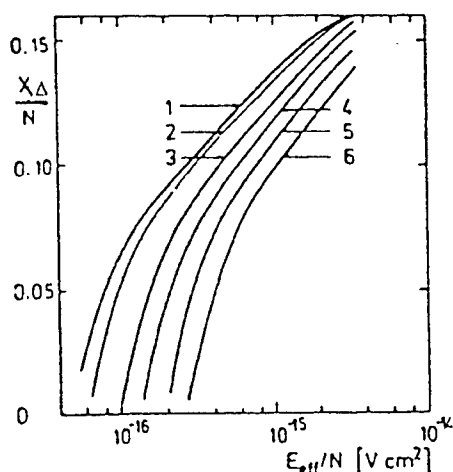


Fig. 2. Calculated relative concentration of the $O_2(^1\Delta_g)$ as a function of E_{eff}/N for several values of the parameter ω/N : 1 – 2.65×10^{-8} ; 2 – 4.7×10^{-8} ; 3 – 9.4×10^{-8} ; 4 – 1.33×10^{-7} ; 5 – 1.88×10^{-7} ; 6 – $2.65 \times 10^{-7} [\text{cm}^3 \text{s}^{-1}]$.

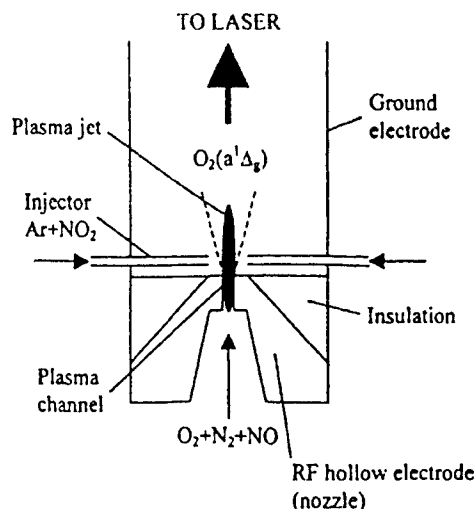


Fig. 3. Radio-frequency plasma jet generator of $O_2(^1\Delta_g)$

3. ATOMIC IODINE GENERATION

A generation of atomic iodine in the ground state, $I(^2P_{3/2})$, directly in the COIL medium to replace its conventional production from dissociated molecular iodine by $O_2(^1\Delta_g)$ opens up a way for potential improvement of the COIL performance. The first is the removal of the $O_2(^1\Delta_g)$, energy to dissociate I_2 , allowing this energy to be extracted as power. The second is the decrease in the molecular weight of the injectant, allowing for faster diffusive mixing, and avoiding negative effect of I_2 on the $I(^2P_{1/2})$ quenching. Also the technical problems induced by special treatment of I_2 vapour could be diminished.

With these needs in mind, our efforts has been focused currently on the gas phase chemical method of atomic iodine generation employing a principal reaction



The reaction (13) represents in fact a multiple gas phase chemical processes that occur in the overall reaction system. It involves initially the generation of F and Cl atoms, respectively, in the reaction of molecular fluorine, F_2 , or chlorine dioxide, ClO_2 , with nitrogen oxide, NO. Additionally, each of these processes is composed of multiple individual chemical reactions describing production and loss processes, including species contained in the COIL medium. And finally, this set of processes occurs within a flowing gas with the exothermicity of the reactions, which brings into the system a complication with coupling the chemical kinetics to the fluid dynamics.

The goals of our work on this problem are: 1/ to provide an overview of the current theoretical understanding of the two complex reaction systems, and to fit them to experimental conditions characteristic for a COIL, 2/ to test experimentally the both reaction systems in a flow reactor (a pilot small-scale device) to prove their suitability for a realistic COIL, and 3/ to test prospectively the developed method of atomic iodine generation directly on an adapted supersonic COIL device built in our laboratory. Work towards the first goal has involved, to date, a model based on the generalized one-

dimensional (1-D) flow development, exploring the chemical processes within the reaction systems. The calculations have been performed for physical conditions (pressure, temperature, and flow rates) applicable to a subsonic region (i.e., upstream of the nozzle throat) of the 1kW-class supersonic COIL operated in our laboratory. The effects of initial concentration ratio of reactants and different configuration of reactants injection were considered. A rate of I atoms generation has been investigated in a flow of nitrogen (as a buffer gas), and a reactive stream containing $O_2(^1\Delta)$. A reaction mechanism for the kinetics of I atoms generation in the $(Cl+HI+O_2(^1\Delta))$ and $(F+HI+O_2(^1\Delta))$, respectively, system has been developed and sensitivities to key processes in this mechanism have been considered. The modeling provided the concentration profiles of all relevant reaction components and the temperature profile along the reaction path. The results of this relatively rough estimation showed that I atoms can be generated via F atoms with the yield of up to 70%, however, on a rather long reaction path (20-40 cm). The reaction system via Cl atoms provided by these calculations I atom yield 80% on substantially shorter reaction path (~4 cm). For this reason, the later system would meet better the COIL mixing conditions. Atomic iodine could be generated near the critical nozzle plane of the supersonic COIL or during the transonic mixing. The chemical kinetics of fluorine system is however simpler, and all reactants are commercially available in gas cylinders. A detailed description of results on the 1-D modeling of both reaction paths for chemical generation of atomic iodine, and their interpretation were partly published^{30,31}, and some advanced results can be found in the paper of this Proceedings.³² A more sophisticated estimation of optimal conditions for I atoms generation needs the 2-D or 3-D modeling, involving also the fluid dynamics effects. This work is planned for a near future.

Work towards the second goal above has involved to date the experimental investigation of I atoms generation via Cl reaction system. A design of the small-scale flow reactor, and the experimental conditions were developed on the basis of the results of the above-mentioned modeling. Preliminary experiments provided atomic iodine with the molar flow rate up to 160 $\mu\text{mol/s}$, and I yield over 70% (in nitrogen).³³ Effects of initial ratio of reactants, and the way of their mixing were studied during the following on investigation. The yields of atomic iodine in the range of 70-100% (related to HI or ClO_2 flow rates) were attained. Some advanced results can be found in the paper of this Proceedings.³⁴ The experiments on atomic iodine generation in the $O_2(^1\Delta)$ stream are under way.

4. REPETITIVELY PULSED SUBSONIC COIL

A period of the early ninetieth signified one of the meaningful millstones in the COIL R&D – a demonstration of ability to operate a COIL in the pulsed regime. A prestige of COIL among other laser systems, particularly technological ones, so has increased. The magnetic gain modulation in a COIL could make possible, e.g., a more efficient non-linear conversion of the wavelength 1.315 μm to the second and higher harmonics. A slow (non-pulsed) control of 3-4 gain could be utilized for a continuous regulation of CW power (within 0-100% of the maximum value) without any change of flow or other operation parameters. Further, it could be employed as a method of the output power stabilization by means of the negative feedback controlling the magnetic field intensity in the gain region.

Besides previously investigated pulsed methods applicable to the COIL, like a pulsed UV photolysis of alkyl iodides in gas containing singlet oxygen^{e.g.35}, and electric discharge initiation of I atoms production from alkyl iodides^{e.g.36}, a Q-switching technique either mechanical³⁷ or magneto-optical³⁸ was also applied to a COIL. The optical switching utilizes the phenomenon of suppressing the gain on the basis of the Zeeman effect.³⁹ Owing to a low gain, low output coupling, and near threshold generation in a COIL, the Q-switching showed a disadvantage due to an increase in intracavity losses.

The pulsed technique investigated in our laboratory is also based on principle of the magnetic suppression (modulation) of gain, but the Zeeman effect is induced by externally applied magnetic field on the gain region.⁴⁰ The small signal gain on the strongest spectral line 3-4 observed in the laser generation in zero magnetic field is weakened in growing magnetic field, and due to above-mentioned physical characteristics of COIL, rather low magnetic fields are needed to quench the laser generation at 1.315 μm . Such a threshold or under-threshold condition can be expressed by the relation⁴¹

$$(2 g_{34}(B) L) / (\delta_0 + \delta_c) \leq 1 \quad (14)$$

$g_{34}(B)$ - small-signal gain at magnetic field B , L - gain length, δ_o - intracavity losses, δ_e - resonator output coupling. Peak power enhancement over cw power can be achieved in the pulsed mode when pulsed power extraction times are short compared to the time that it takes to fill the outcoupled gain volume with fresh "fuel" (in a subsonic system usually $\sim 10^{-3}$ s). Energy of the single pulse is given by the average CW power (with zero magnetic field), and the optimal repetition rate. A pulsed length and its enhancement over CW power depend also on a switching off rate of the magnetic field. The gain volume, from which power can be extracted, is limited by deactivation rate of both the singlet oxygen and excited atomic iodine. Provided that this rate is fixed for a particular temperature in the cavity region, the way to increase this volume and thus increase the peak power enhancement is to increase the velocity of the flowing laser gases, and so to increase the concentrations of $O_2(^1\Delta_g)$ or atomic iodine or both. Experimental verification of the proposed pulsed technique was performed on the small-scale subsonic COIL device generating the CW output power up to several tenth watts. It was modified to apply magnetic field to the gain cavity from two rectangular coils attached to the laser body made of Plexiglas (see Fig. 4).

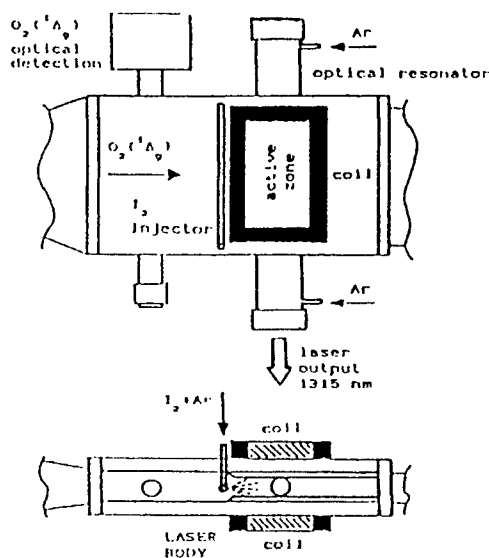


Fig. 4. Scheme of set up for magnetic pulsed regime of COIL

A typical oscillogram of the periodically pulsed COIL is shown in Fig. 5 that was recorded for 15 W of the average CW power. The peak enhancement corresponded to the power of 48 W, i.e., the enhancement ratio was 3.2. A length of single pulse was ~ 200 μ s at the rate of magnetic filed pulsing of 0.5 G/ μ s.⁴¹ A maximum peak power enhancement achieved on this experimental configuration was ~ 3.5 only with the repetition rate of ~ 1 kHz. It was given by the relatively long switching off time of the used magnetic field generator. Consequently, extraction of the pulse power could not be fully developed.

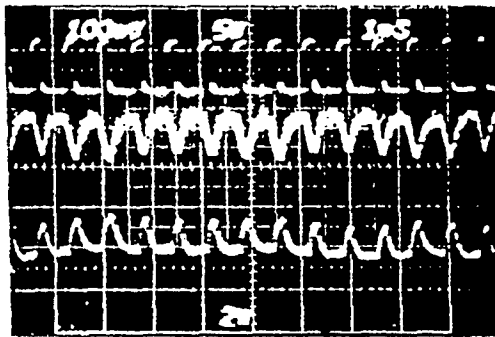


Fig. 5. Oscillogram of the repetitively pulsed COIL.
upper trace – pulsed magnetic filed (1 ms/div, peak current 250 A);
lower trace – laser pulsed power (CW power 15 W, pulsed power 48 W)

Concurrently with our work, the magnetic field nullified-switched gain technique was developed at the AFRL by Hager *et al.*⁴² This technique works on principle of the gain suppressing below the lasing threshold by a static magnetic field that is followed by applying a fast rising current pulse to cancel the cavity magnetic field. A switching off the magnetic field was very fast in this way ($\sim 0.5 \mu\text{s}$) resulting in the laser pulse extraction with duration less than $20 \mu\text{s}$, and a peak-CW power enhancement ratio of 12.7.

Experimental data on estimation of the "3-4 gain quenching" magnetic field intensity, called a threshold magnetic field, were also collected during this investigation. It is defined as $g(B_{th}) = g_{th}(B=0)$.⁴³ The magnetic field was continuously increased up to level when the output laser power quenched. The values of $g(B_{th})$ evaluated in dependence on iodine molar flow rate in the cavity, and for several values of output coupling parameter are plotted in Fig. 6. This dependence showed a linear character in the overall tested parameters, even though the dependence of laser power, as well as output coupling, on iodine molar flow rate, showed generally maximum for a certain value of iodine concentration in the cavity.

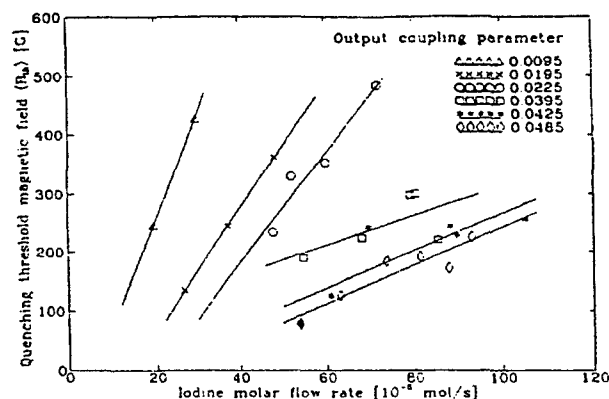


Fig. 6. Dependence of threshold magnetic field on iodine molar flow rate for different output couplings

It can be concluded from these results that a certain value of threshold magnetic field is affected by a set of physical and operation parameters together, particularly by iodine molar flow rates, small signal gain, threshold gain, and output couplings. In spite of the fact that a mutual relation among these parameters is rather complex, we proposed using a dimensionless parameter, the normalized gain defined as the ratio of threshold and small-signal gain (g_{th}/g), to evaluate from the experimental data the threshold magnetic field for given conditions. The threshold gain includes the effect of output coupling, intra-resonator losses, and gain length. Iodine flow rate, Doppler and pressure broadening of the laser transition, and also mixing efficiency affect the small signal gain. The dependence of normalized gain on the threshold magnetic field is shown in Fig 7.

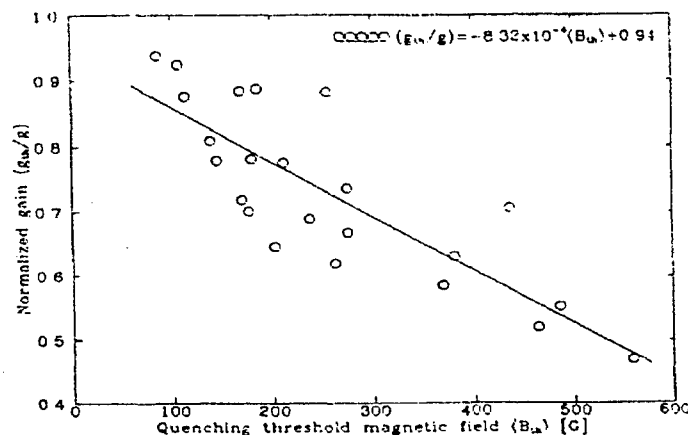


Fig. 7. Normalized gain (g_{th}/g_0) in relation to threshold magnetic field

5. OPERATION OF 1 kW-CLASS SUPERSONIC COIL

5.1. Performance characteristics

A small-scale 5-cm gain length supersonic chemical oxygen iodine laser driven by a jet singlet oxygen generator (jet SOG) with a closed-loop liquid flow system has been developed since 1996, and laser characteristics have been investigated. Parameters of the principal device parts are similar to the VertiCOIL developed in the AFRL except the rotating disks generator. The PC on-line with designed hardware and software nearly fully automatically controls the all system and records 32 essential laser parameters. An iodine vapour in helium is injected into the primary oxygen/helium flow in a subsonic region 5 to 7 mm upstream from the nozzle throat plane. A single horizontal slit configuration of nozzle is used for the adiabatic expansion of gas flow. A distance between the sonic plane and optical axis of the laser resonator is adjusted from 35 to 55 mm. A stable configuration of the resonator with 5 cm-diameter inner mirrors is employed. The mirrors are protected by helium atmosphere. The nozzle ramps and mirrors holders limit the multimode output beam size to 3.7 cm in the flow direction by 1.5 cm between the ramps.

Few experimental results are shown to represent state of the art of the investigation performed on this device to date. The jet SOG is operated typically at pressure 7.5 to 9.0 kPa (60 to 70 torr), with helium admixed to chlorine in the ratio mostly 1:2. A pressure in the generator, and consequently $O_2(^1\Delta_g)$ yield could be easily controlled by throttling the gas at the generator exit. It can be seen from one example of the generator performance record shown in Fig. 8. An increase in the yield with pressure decreasing results from a shorter residence time of gas in the generator, and also a lower $O_2(^1\Delta_g)$ partial pressure. Both effects diminish $O_2(^1\Delta_g)$ loss in the self-quenching/pooling reactions¹¹

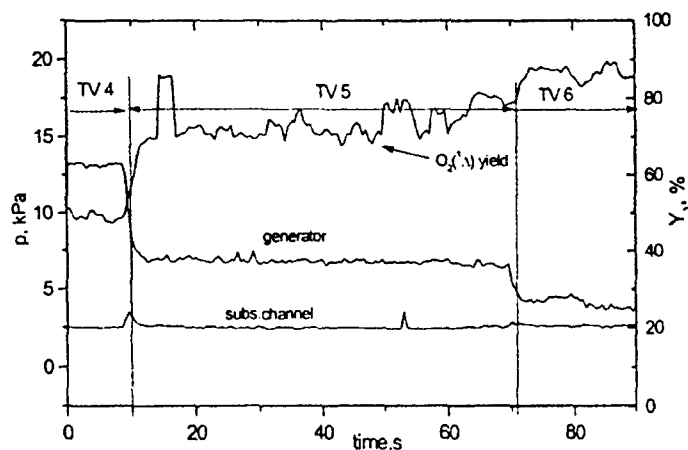
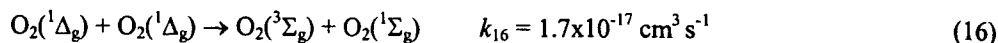
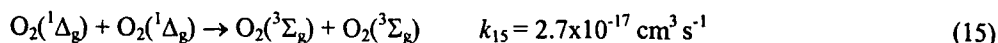


Fig. 8. Pressure in jet generator and $O_2(^1\Delta_g)$ yield controlled by gas throttling at generator exit. Position of throttle valve flap in tens of degrees is denoted by TV numbers (TV0 - closed, TV9 - opened entirely). Cl_2 flow rate of 40 mmol/s, He_{prim} flow rate of 40 mmol /s.

The jet SOG driving the supersonic COIL provided typically rather high $O_2(^1\Delta_g)$ yields (see Table1).

Tab. 1

P_{gen} , kPa	7	8	9	10
$O_2(^1\Delta_g)$ yield, %	80	77	70	63

Gas dynamic conditions in the whole laser system were tested during a „cold flow run“, i.e., with gaseous nitrogen instead of chlorine was introduced into the primary flow. The average Mach number, M_1 , for a subsonic flow ahead of the sonic throat, and the supersonic flow in the resonator region, M_2 , were evaluated from pressure ($P_{1,2}$) in the respective regions of the device, gas molar flow rates, (n), physical properties of the flowing gas (molecular weight, MW , and adiabatic constant, κ), stagnation temperature, and the flow cross section.⁴⁴ The local Mach number, M_2' , in the supersonic region was evaluated from the pressure ratio P_{stat}/P_{Pit} (P_{stat} - static pressure measured at the cavity wall, P_{Pit} - stagnation pressure measured by the Pitot tube in the cavity centreline placed 55 mm from sonic throat).⁴⁵ The results are summarized in Table 2.

Tab. 2

n_{N_2} , mmol/s	n_{He} , mmol/s	P_1 , Pa	P_2 , Pa	P_{Pit} , Pa	MW , kg/mol	κ	M_1	M_2	M_2'
21	90	2055	174	1457	8.6×10^{-3}	1.57	0.41	1.94	2.37

Laser output power was investigated at various iodine flow rates and output couplings. One example of such dependence is shown in Fig. 9. A simplified Rigrod theory⁴⁶ for saturation intensity was employed to estimate the small signal gain and saturation intensity from these experimental data. The gain of $1.29 \times 10^{-2} \text{ cm}^{-1}$, and the saturation intensity of 5.4 kW/cm^2 were evaluated.

The effect of titration ratio $[I_2]/[O_2(^1\Delta_g)]$ on laser power was searched also for different output couplings. Fig. 10 illustrates it for two values of generator pressure. The highest power was mostly achieved for $[I_2]/[O_2(^1\Delta_g)] = (5 \pm 1)\%$. The optimum value of 1.7% obtained on the VertiCOIL system⁴⁷ was significantly lower, however this discrepancy diminishes to 3.1% when the titration ratio is recalculated by dividing the total $[O_2]$ in VertiCOIL with $O_2(^1\Delta_g)$ yield (0.55).

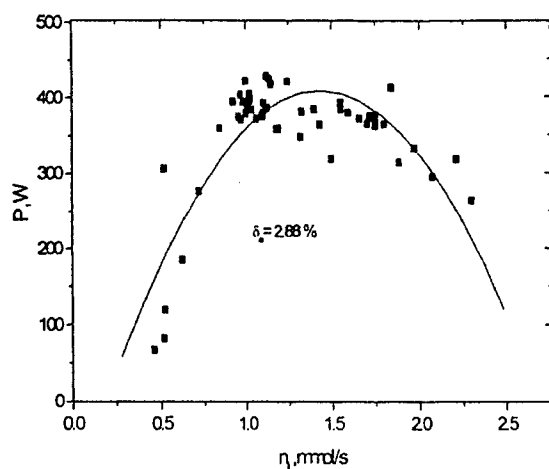


Fig. 9. Laser power as a function of iodine flow rate. Output coupling 2.83% ($T_1 + T_2$), 40 mmol Cl_2/s , 80 mmol He_{prim}/s , 40 mmol He_{sec}/s .

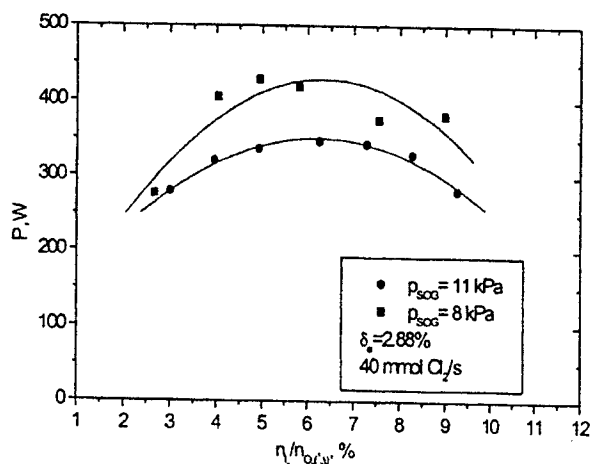


Fig. 10. Laser power as a function of titration ratio $[I_2]/[O_2(^1\Delta_g)]$ at two pressures in generator

The optimum dilution ratio, $[He_{sec}]/[I_2]$, at maximum attainable laser power was found in the range from 1:30 to 1:70. These measurements were directed to get efficient mixing of the primary gas flow, $O_2(^1\Delta_g) + He_{prim}$, and the secondary gas flow, $I_2 + He_{sec}$. It was estimated by means of the relative penetration factor, $\pi_{rel} = \pi/\pi_{full}$. The penetration parameters, π and π_{full} , describing the mixing both gas flows are evaluated from their physical properties, and hardware parameters of the device.⁴⁸

$$\pi = (n_s/n_p) [(M_s T_s P_p) / (M_p T_p P_s)]^{1/2} \quad (17)$$

$$\pi_{full} = (d A_s) / 5 D A_p \quad (18)$$

n - flow rate, M - molecular weight of gas mixture, T - temperature, P - pressure, d - height of primary flow channel, A - flow cross-section, D - orifice diameter of secondary flow (index p – primary flow, index s – secondary flow). The best mixing conditions resulting in the maximum output power were at the values of π_{rel} between 0.8-1.0.

State of the art of the performance characteristics of the COIL device operated in our laboratory are summarized in Table 3. The laser operation has not been assessed as enough efficient yet. The reason for a rather low chemical efficiency (measured output power) is most probably a bad quality of the resonator mirrors used so far in this laser system.

Tab. 3.

Laser power	430 W
Running time	1.5 min
Chlorine flowrate	37.8 mmol/s
Primary helium diluent	80 mmol/s
Generator pressure	8 kPa (60 Torr)
Chlorine utilization	0.97
Starting BHP molarity	6.7 M HO ₂ ⁻
O ₂ (¹ Δ _g) measured yield	0.72
Pressure upstream the nozzle	3.8 kPa (28.5 Torr)
Laser cavity pressure	380 Pa (2.8 Torr)
I ₂ /O ₂ flow rate ratio	0.029
Mirrors reflectivity	0.9995/0.981
Mode length	3.7 cm
Small signal gain (Rigrod)	0.013 cm ⁻¹
Saturation intensity	5.4 kW/cm ²
Chemical efficiency	0.12

5.2. Small signal gain distribution and cavity temperature evaluation

A spatial distribution of the small signal gain, and evaluation of cavity temperature was investigated by means of the Iodine Scan Diode Probe diagnostics developed for a COIL at the PSI⁴⁹, and used first at the AFRL.⁵⁰ The line-shape of the COIL gain profile was scanned in frequency by a diode laser of narrow line width operating in the region of the COIL transition frequency. The small signal gain, $g(\nu_0)$, was determined using the equation⁴⁷

$$I(\nu_0) = I_0(\nu_0) \exp(g(\nu_0) L) \quad (19)$$

$I_0(\nu_0)$ - line-center probe laser intensity entering the gain region, $I(\nu_0)$ - line-center intensity after amplification through the gain region, L - gain length.

The temperature in terms of the Doppler full width at half-maximum, $\Delta\nu_D$, was evaluated from the relation⁴⁷

$$\Delta\nu_D(T) = 2 \nu_0 / c \{ \ln 2 (2 k T / M) \}^{1/2} \quad (20)$$

ν_0 - central frequency, c - light velocity within medium, k - the Boltzmann's constant, M - mass of medium species.

Performed mapping of the gain distribution through the gain region was aimed at the inspection of mixing conditions in the cavity at different experimental conditions, and bearing out our guess at the reason of rather low chemical efficiency of this device (mirrors quality). The results presented below were gathered for the primary gas flow of 40 mmol Cl₂/s

and 80 mmol $\text{He}_{\text{prim}}/\text{s}$, the secondary gas flow of 40-80 $\text{He}_{\text{sec}}/\text{s}$ and 1-2 mmol I_2/s . The plenum pressure was 3.4 - 4 kPa (25-30 Torr), the cavity pressure measured at the upper wall in the location of the resonator axis was 300-390 Pa (2.2-2.8 torr). The 2.5 mm i.d.-beam of the probe laser was passed through the gain medium parallel to the optical axis. A two-pass configuration of the probe beam was chosen for the data extraction. The probe beam emitter/detector unit and the mirror system for beam alignment were mounted on an assembly of the motorized linear positioning equipment that allowed fast and precise movements either in horizontal direction (x axis) along the gas flow or vertical direction (y axis) perpendicular to the gas flow. The grid of measuring positions is presented in Fig. 11.

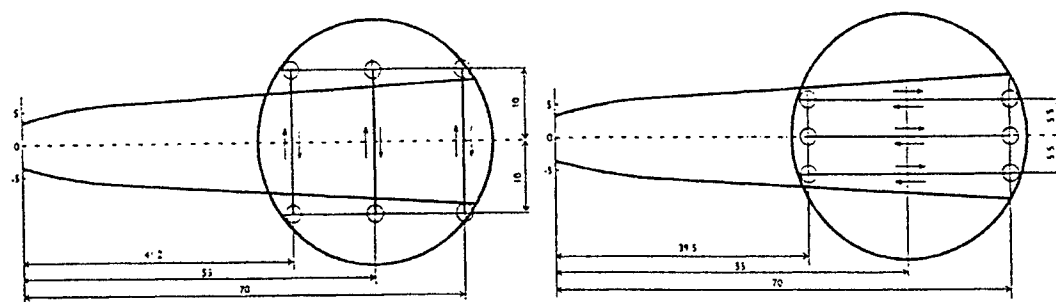


Fig. 11. Scheme of probe beam scanning in vertical and horizontal position

Duration of each scan was 13.3 seconds, and the gain data were recorded within 60-100 seconds' hot runs. One example of the gain distribution curves cross the gain region in the vertical direction measured at three values of I_2 flow rate is illustrated in Fig. 12. A parabolic shape of the gain distribution curve indicates that the gas flow velocity across the gain region was inhomogeneous resulting in a slower gas flow at boundary layers (0.5 to 1.5 mm thick) along the cavity walls. In these layers, a remarkable quenching of excited I atoms occurred. Molecular iodine may be also present in these layers which may consume $\text{O}_2(^1\Delta_g)$ in its dissociation. The maximum gain was attained at the iodine flow rate of 1.5 mmol I_2/s . Similar parabolic distribution of gain was obtained for different distance from the nozzle, but the maximum gain values were attained at different iodine flow rates under constant other conditions. An insufficient mixing of primary and secondary flows (inefficient penetration) could be easily detected by this diagnostics (see Fig. 13). The gain distribution record in this case (40 mmol $\text{He}_{\text{sec}}/\text{s}$ only) showed two peaks and minimum located in the cavity center, which can be explained by a shortage of iodine is in the center of the cavity.

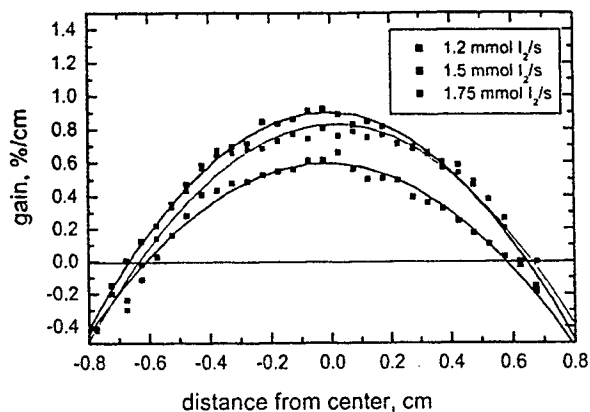


Fig. 12. Gain distribution in vertical direction, 5.5 cm downstream nozzle throat (resonator axis); for three I_2 flow rates; 80 mmol $\text{He}_{\text{sec}}/\text{s}$.

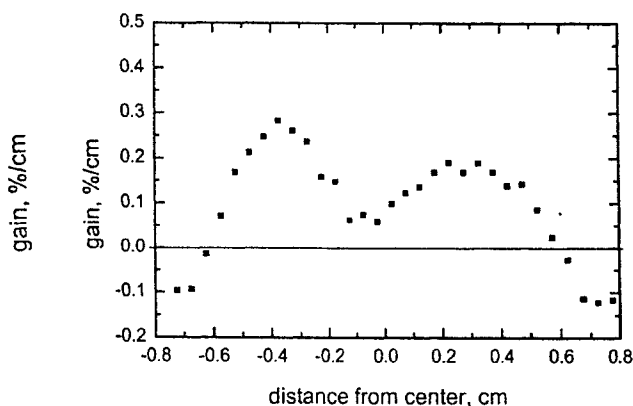


Fig. 13. Gain distribution in vertical direction, 5.5 cm downstream nozzle throat (resonator axis); 1.5 mmol I_2/s , 40 mmol $\text{He}_{\text{sec}}/\text{s}$.

A plot of measured gain on evaluated relative penetration factor is shown in Fig. 14. The small signal gain of ~1%/cm was typical for the supersonic COIL device operated in our laboratory at optimal mixing conditions. Comparing it, for

example, with the gain attainable on the RADICL device (~ 1.2 %/cm), and considering also twice higher cavity pressure in this laser system (5.8 Torr), there is a prospect to enhance the gain of our COIL driven by jet SOG when the cavity pressure is increased.

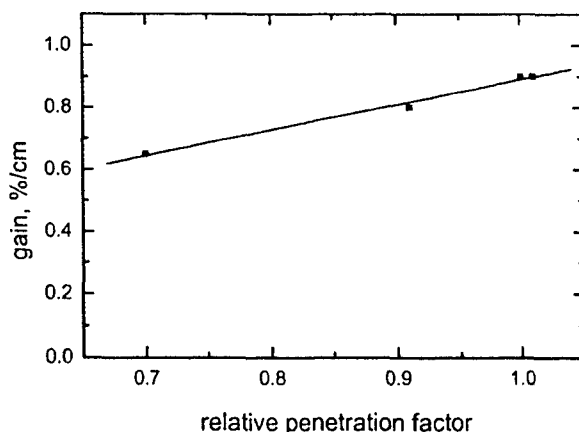


Fig. 14. Gain in the cavity center in relation to calculated relative penetration factor; 5.5 cm from the nozzle throat (resonator axis), 1.5 mmol I_2/s , 80 mmol H_{2O}/s .

The gas temperature calculated from these measurements was not homogeneous across the whole cavity. In the boundary layers when the probe beam recorded weak emission or even absorption, the gas temperature was much higher (>350 K) than in the cavity center (175 – 205 K). A low gas velocity and high rate of exothermic processes per unit length in the boundary layers near the walls to the cavity center can explain it.

6. SUMMARY

The above-presented results have been extracted from a number of publications and reports that have come out from the COIL Laboratory in Prague. We hope that our current and future research on COIL related problems contributes and will contribute to the advanced COIL technology.

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