A History of Coil Development in the USA

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ABSTRACT

This is an overview of the development of Chemical Oxygen-Iodine Laser (COIL) technology in the United States. Key technical developments will be reviewed, beginning in 1960 and culminating in 1977 with the first COIL lasing demonstration at the Air Force Weapons Laboratory [now the Phillips Laboratory]. The discussion will then turn to subsonic laser development, supersonic lasing demonstration and efficiency improvements, and finishing with a brief discussion of some spin off COIL technologies. Particular emphasis will be placed on how the O_2 ($^1\Delta$) generator and O_2 - I_2 mixing nozzle technologies evolved.

1. INTRODUCTION

The development of the COIL laser over the past 16 years is a remarkable achievement. The energy source, a chemical reaction between gaseous chlorine and aqueous basic hydrogen peroxide (BHP), is extraordinarily specific, producing 100% of the oxygen in the $^1\Delta$ state. The electronically excited $O_2(^1\Delta)$ is then used to dissociate a small amount of $I_2(X)$ [$I_2/O_2 \le 0.04$]. The laser energy, which is stored in the $O_2(^1\Delta)$, is then transferred to the iodine atoms (3);

$$I(^{2}P_{3/2}) + O_{2}(^{1}\Delta) \rightarrow I(^{2}P_{1/2}) + O_{2}(^{3}\Sigma)$$

 $\Delta H_{Ia} = -279cm^{-1}$ (1a)

$$k_{la} = 7.6X10^{-11} cm^3 / molecule / sec$$

 $K_{eq} = 0.75 Exp[402 / T]$ (1b)

and lasing occurs at 1.315 µm;

$$I(^{2}P_{1/2}) + nh\nu_{ir} \rightarrow I(^{2}P_{3/2}) + (n+1)h\nu_{ir}$$
 (2)

At present CO_2 and Nd:YAG lasers form the industrial base for laser machining and treatment (cutting, welding, drilling, surface treatment, etc.). The Nd:YAG lasers are expensive to operate and CO_2 laser wavelength (10.6 μ m) couple poorly into metals⁽⁴⁾. The COIL laser can operate in the cw or pulsed mode, the beam quality is inherently very good which insures narrow beam divergences, the transmission of 1.315 μ m radiation through optical fibers is excellent, and the inexpensive chemicals (KOH, H_2O_2 , Cl_2) make the laser a good candidate for industrial application. This paper will detail the development of COIL technology in the United States and consider the potential for industrial development.

2. FUNDAMENTAL DEVELOPMENTS LEADING TO COIL (1960-1978)

The fundamental work that lead to the COIL was published over the 18 year period between 1960 and 1978. The essential demonstrations were: (1) use of chemical reactions to produce population inversions, (2) transfer of energy from "Hot" chemical reaction products to "Cold" lasing species, (3) lasing on the $I(^2P_{1/2})$ - $I(^2P_{3/2})$ atomic transition, (4) development of $O_2(^1\Delta)$ production methods, and (5) recognition that the near resonant energy transfer reaction in Eq. 1 could produce population inversion and support lasing.

In the 1961 paper by Polanyi⁽⁵⁾ on chemical lasers he suggested that the vibrationally excited product molecules from chemical reactions could support lasing at IR wavelengths. In 1965 Kasper & Pimentel demonstrated lasing on HCl vibrational transitions from flash lamp initiated reaction of H_2/Cl_2 mixtures and in 1967 Deutsch and Kompa & Pimentel⁽⁸⁾ demonstrated pulsed HF lasing. These experiments confirmed that chemical reactions could be used to produce population inversion and lasing. In 1969 Cool & Stephens⁽⁹⁾ experimentally showed that a vibrational transfer laser was feasible by

lasing vibrationally excited CO_2 pumped with chemically produced vibrationally excited DF molecules. This was the first demonstration of a pure CW chemical laser.

The first atomic transition laser was reported in 1964 when Kasper & Pimentel⁽¹⁰⁾ operated a photo dissociation iodine laser;

$$CX_3I + hv_{uv} \to CX_3 + I(^2P_{1/2})[X = H, F]$$
 (3)

The electronically excited $I(^2P_{1/2})$ then lases (see Eq. 2). These experiments set the stage for developing the COIL laser.

Development of chemical $O_2(^1\Delta)$ generators between 1960 and 1979 can be attributed to a number of researchers; Seliger^(11, 12), Kahn & Kasha⁽¹³⁻¹⁶⁾, Held et. al.⁽¹⁷⁾, and McDermott and Benard^(1, 18). Kerns⁽¹⁹⁾ published an excellent review in 1971.

BHP is the primary fuel for COIL and is produced by mixing an alkali metal hydroxide, usually NaOH or KOH, with hydrogen peroxide:

$$H_2O_2 + MOH \rightarrow M^+ + O_2H^- + H_2O$$

$$(excess)$$

$$[M = Na, K]$$

$$k_4 = 4 \times 10^4$$

$$\Delta H_4 = -12 kcal / mole$$
(4)

This equation shows that BHP is composed of O_2H^- , H_2O_2 , and H_2O with essentially no OH $^-$. The standard BHP solutions used at the Phillips Laboratory are (7-8) molar in O_2H^- , (1-3) molar in H_2O_2 , and about 50% by weight H_2O . Excess H_2O_2 in the BHP is important to avoid excess heat generation from the reaction of Cl_2 with OH $^-$ [excess heat is defined here as heat generation that does not lead to $O_2(^1\Delta)$ production]. Singlet delta oxygen is then produced via the exothermic reaction between chlorine and BHP, a reaction that is postulated to have three steps⁽¹⁷⁾, Eqs. 5-7 with the overall reaction shown in Eq. 8.

$$O_2H^- + Cl_2 \rightarrow HOOCl + Cl^-$$

$$k_5 = 2.7 \times 10^{10} \, Cm^3 / molecule / sec$$
(5)

$$O_2H^- + HOOCI \rightarrow CIO_2^- + H_2O_2$$

$$k_6 = \infty$$
(6)

$$ClO_2^- \to Cl^- + O_2(^1\Delta)$$

$$k_7 \ge 500 / \sec$$
(7)

$$Cl_2 + 2O_2H^- \rightarrow O_2(^1\Delta) + 2Cl^-$$

$$\Delta H_8 = -27 \, kcal \, / \, mole$$
(8)

Derwent & Thrush⁽³⁾ were the first to recognize that the nearly resonant energy transfer reaction between $O_2(^1\Delta)$ and atomic iodine, see Eq. (1), could be used to support atomic iodine lasing [Figure 1 shows the energy level diagram]. Although the first attempts failed^(20,21), McDermott et. al. reported success in 1978. Benard et. al.⁽¹⁸⁾ and Richardson et. al.⁽²²⁾ quickly published papers that established the basic elements for all subsonic COIL lasers to follow.

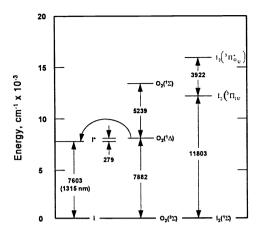


Figure 1. Chemical Oxygen-Iodine Laser Electronic Energy Level Diagram

3. PHASE OF COIL DEVELOPMENT IN THE USA (1977-PRESENT)

COIL development in the United States can be divided into four phases (see Table 1); (1) subsonic COIL development (1977-1984), (2) supersonic COIL lasing demonstration (1982-1984), (3) COIL engineering demonstrations (1984-1989), and (4) COIL efficiency improvements (1990-Present). During this evolution of COIL laser technology several spin off technologies have also been developed. These include frequency doubling and magnetic gain switching (24, 25). All of these topics will be discussed in the following sections.

Table 1. COIL History in the USA. AFWL=Air Force Weapons Laboratory (now the Phillips Laboratory=PL)
KAFB, NM; McD=McDonnell Douglas, Kansas City, MO; TRW=TRW, Redondo Beach, CA; RD=Rockwell Corp.,
Rocketdyne Division, Canoga Park, CA

Year	Reference	Location	Cl ₂ Flow (moles/sec)	Power (Watts)	Delta (W/cm ²)	Efficiency
			Phase I	(watts)	(W/CIII)	
1977	McDermott	USA (AFWL)	0.004	.004	2 x 10 ⁻⁵	1 x 10 ⁻⁵
1978	Benard	USA (AFWL)	0.03	100	0.8	0.037
1979	Richardson	USA (McD)	0.015	10	0.081	0.007
1981	Berg	USA (TRW)	0.2	2000	6.7	0.11
1981	Truesdell	USA (RD)	0.05	150	0.91	0.033
1982	Hager	USA (AFWL)	0.6	4600	4.6	0.084
1984	Wiswall	USA (McD)	0.02	120	0.067	0.066
1985	Wiswall	USA (McD)	0.02	180	0.1	0.10
			Phase II			
1984	Hager	USA (AFWL)	0.15	1600	40	0.12
1984	Berg	USA (TRW)	0.3	4200	84	0.15
			Phase III			
1985	Dickerson		0.15	1900	58	0.14
1987	Truesdell	USA (AFWL)	1.55	25000	110	0.18
1988	Truesdell	USA (AFWL)	1.55	33000	145	0.23
1989	Truesdell	USA (AFWL)	1.8	39000	172	0.24
1991	Truesdell	USA (PL)	0.6	10000	200	0.18
			Phase IV			
1993	Helms	USA (PL)	0.035	430	153	0.14

3.1 Phase I: Subsonic COIL Development (1977-1984)

After the initial scaling of COIL from the milliwatts level to 100 watts by Bernard et. al. (18), the subsonic COIL was further scaled to 2 kW (26), and then to 4.6 (27), using exact "sewer pipe quantum engineering" (28). During these experiments the chemical physics of $O_2(^1\Delta)$ production was not well understood, nor were the gas phase kinetic processes involved in $I(^2P_{3/2})$ pumping, and $I(^2P_{1/2})$ deactivation and lasing. Fortunately the forgiving nature of COIL helped lead to success and the key technical achievements were; (1) an increased understanding of the I_2 dissociation process, (2) $O_2(^1\Delta)$ generator improvements, and (3) oxygen-iodine mixing nozzle development. Intimately linked to these developments was an improved understanding of the major COIL loss mechanisms.

3.1.1 I₂ Dissociation

During the late 1970's and early 1980's many investigators worked on the dissociation of I_2 by $O_2(^1\Delta)$ and an excellent review is presented by Heidner et. al. (29, 30). Heidner's proposed auto-catalytic chain mechanism for I_2 dissociation is initiated by one or a combination of four reactions:

$$O(^{3}P) + I_{2}(X) \rightarrow I(^{2}P_{3/2}) + IO$$
 (9)

$$O_2(^1\Sigma) + I_2(X) \rightarrow 2I(^2P_{3/2}) + O_2(^3\Sigma)$$

 $k_{10} = 10^{-11} \text{ cm}^3 / \text{ molecule / sec}$
 $\Delta H_{10} = 1.6 \text{ kcal / mole}$ (10)

$$O_2(^1\Delta) + I_2(X) \to I_2^*(X) + O_2(^3\Sigma)$$

 $k_{11} = 7 \times 10^{-15} \text{ cm}^3 / \text{molecule / sec}$ (11)

$$I_2^*(X) + O_2(^1\Delta) \rightarrow 2I(^2P_{3/2}) + O_2(^3\Sigma)$$

$$[3 \times 10^{-11} < k_{12} < 3 \times 10^{-10}$$

$$cm^3 / molecule / sec]$$
(12)

and the $I(^2P_{3/2})$ atoms formed are pumped to $I(^2P_{1/2})$ via Eq. 1. Chain branching occurs by Eq. 11 or when $I_2(X)$ collides with $I(^2P_{1/2})$ to form $I_2*(X)$;

$$I(^{2}P_{1/2}) + I_{2}(X) \to I_{2}^{*}(X) + I(^{2}P_{3/2})$$

$$k_{13} = 3.5 X \cdot 10^{-11} cm^{3} / molecule / sec$$
(13)

The $I_2^*(X)$ formed in Eqs. 11 and 13 then dissociates into $I(^2P_{3/2})$ atoms upon collision with $O_2(^1\Delta)$ [Eq. 12] or collision with $I(^2P_{1/2})$:

$$I_2^*(X) + I(^2P_{1/2}) \to 3I(^2P_{3/2})$$
 (14)

Although the iodine intermediate $I_2^*(X)$ has not been observed directly the evidence strongly suggests it is vibrationally excited $I_2(X)$ and not one of the low lying triplet $I_2(A)$ electronic states^(30, 31).

These investigations lead to the idea that initiation of the I_2 dissociation in the high pressure nozzle plenum may be preferable to supersonic injection and mixing of I_2 with $O_2(^1\Delta)^{(32)^2}$.

3.1.2 Major COIL Loss Mechanism

Singlet oxygen pooling and wall deactivation determine the maximum $O_2(^1\Delta)$ that can be delivered to the COIL laser cavity⁽²⁹⁾;

$$O_2(^{1}\Delta) + O_2(^{1}\Delta) \rightarrow O_2(^{3}\Sigma) + O_2(^{1}\Sigma)$$

 $k_{15a} = 2 \times 10^{-17} \text{ cm}^{3} / \text{molecule / sec}$
 $\Delta H_{15a} = -8.9 \text{ kcal / mole}$ (15a)

$$O_2(^{1}\Sigma) + M \rightarrow O_2(^{3}\Sigma) + M$$

$$k_{15b} = 10^{-2} cm^{3} / molecule / sec$$

$$\Delta H_{15b} = -15kcal / mole$$
(15b)

$$O_2(^1\Delta) + M \rightarrow O_2(^3\Sigma) + M$$

$$k_{15c} = 2 X 10^{-5} cm^3 / molecule / sec$$

$$\Delta H_{15c} = -22 kcal / mole$$
(15c)

In smaller devices Eq. 15c is important and in larger lasers Eqs. 15a and 15b dominate the loss mechanism.

The second major loss in COIL is caused by water deactivation of both the lasing species $[I(^2P_{3/2})]$ and the I_2 dissociation intermediate $[I_2(X)]$:

$$I(^{2}P_{1/2}) + H_{2}O \rightarrow I(^{2}P_{1/2}) + H_{2}O$$

 $k_{16} = 1.7 \times 10^{-12} \text{ cm}^{3} / \text{molecule / sec}$
 $\Delta H_{16} = -22 \text{ kcal / mole}$ (16)

$$I_2^*(X) + H_2O \rightarrow I_2(X) + H_2O$$

 $k_{17} = 3 X \cdot 10^{-10} \text{ cm}^3 / \text{molecule / sec}$ (17)

3.1.3 Sparger $O_2(^1\Delta)$ Generators

The first $O_2(^1\Delta)$ generators used for COIL lasers were chemical sparger types [see Figure 2] where chlorine gas is bubbled through a column of BHP. The efficiency of these generators depends on the height of the liquid column above the Cl_2 injection, the residence time of the Cl_2/O_2 in the generator gas bubbles, the volume of the transport ducts and the temperature of the bulk BHP. The $O_2(^1\Delta)$ yield depends on the Cl_2 injector hole size and depth below the BHP solution^(26, 27, 28). Proper adjustment of these parameters will result in chlorine utilization near 100% [see Figure 3]. Performance is also affected by the presence of diluent gases such as helium and optimum generator performance occurs at lower "bubble" residence times and lower Cl_2 injection depths as the He: Cl_2 ratio is reduced.

The measured $O_2(^1\Delta)$ yield existing sparger reactors operating in the torr pressure range is near $50\%^{(34)}$. The losses that contribute to this yield are; (1) liquid phase losses (about 0.04 yield points), (2) $O_2(^1\Delta)$ pooling loss in the gas "bubble" (about 0.36 yield points), (3) generator wall quenching losses (about 0.03 yield points), and (4) duct transport loss (about 0.06 yield points). Dimole emission losses are negligible because of the weakness of the transition $[10^4$ to 10^5 times lower than $O_2(^1\Delta)$ pooling].

Another factor that contributes to the efficiency of these generators is the bulk BHP temperature. The lower the temperature, the lower the H_2O partial pressure leaving the generator. Operating at lower temperatures can be accomplished by lowering the solution freezing point with increased concentrations of ions⁽³⁵⁾. This has been achieved by switching from sodium BHP (NaOH + H_2O_2) with a freezing point of -260 K for a 3.5 molar solution to potassium BHP (KOH + H_2O_2) with a freezing point of 230 K for 8 molar solutions .

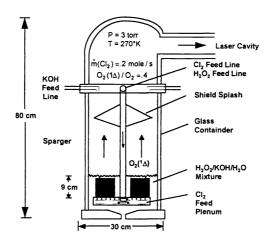


Figure 2. Sparger or Bubbler Type $O_2(^1\Delta)$ Generator

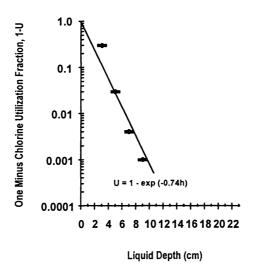


Figure 3. Chlorine Utilization as a Function of Chlorine Injector Depth for the COIL IV Sparger (Ref. 27)

3.1.4 Subsonic COIL O₂-I₂ Mixing Nozzles

Based on early results^(1, 18, 22) it was believed that I_2 dissociation would be rapid, $I(^2P_{1/2})$ deactivation would be minimal, and cavity gain would hold up for tens of centimeters, even at low subsonic velocities projected for COIL $IV^{(38)}$. Consequently early subsonic mixing nozzles were coarse [see Figure 4a] and the laser performance was very poor [the SSG, iodine dissociation, and power varied quite substantially and unpredictably]⁽²⁷⁾. These results can be explained by the poor diffusion of the secondary jets into the primary flow [see the laser induced fluorescence (LIF) data in Figure 5]. A series of sub-scale LIF investigations of the dynamics of jet expansion, pressure matching, and diffusion mixing scale [see Figure 6] was undertaken^(27, 39), resulting in a new injector with more, smaller diameter holes situated perpendicular and parallel to the flow direction [see Figure 4c]. To obtain "good mixing" with these nozzles, secondary to primary flow ratios approaching one were required. The higher relative secondary flows throttled the primary flow, increased the system pressures, and increased the $O_2(^1\Delta)$ pooling losses⁽²⁷⁾. This effect required a compromise between good mixing [high secondary flow] and high generator efficiency [lower secondary flow].

Further optimization of subsonic mixing nozzles was not attempted in the United States after running these COIL IV experiments. At this point the emphasis in COIL research turned to supersonic COIL laser.

3.2 Phase II: Supersonic COIL Lasing Demonstration (1982-1984)

Supersonic COIL development was motivated for three reasons; first to reduce the size of the device, second to lower the cavity operating temperature [increasing the device efficiency], and third to stretch the stream wise gain zone [reducing the density gradients which degrade beam quality].

The size reduction offered by supersonic operation is illustrated in Figure 7 which shows COIL IV, a 4 meter long subsonic COIL laser, and the 25 cm long nozzle from ReCOIL, the first supersonic COIL (32) [both lasers were comparable in power, see Table 1].

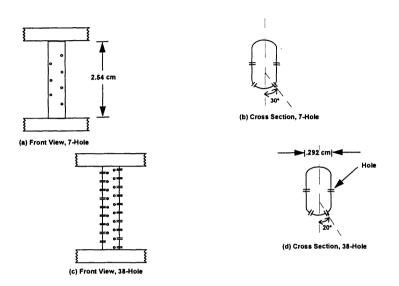


Figure 4. COIL IV 7-Hole and 38-Hole Iodine Injector (Ref. 27)

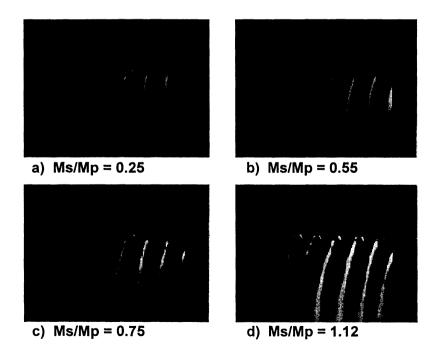


Figure 5. COIL IV 7 Hole injector LIF Photographs (Ref. 27)

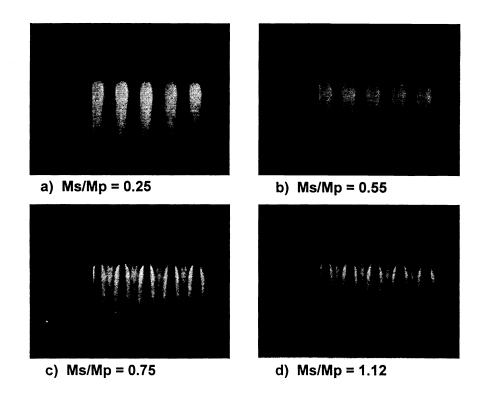


Figure 6. COIL IV 38 Hole Injector LIF Photographs (Ref. 27)

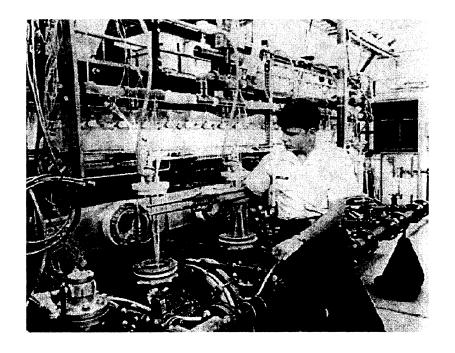


Figure 7. Comparison of COIL IV Subsonic Laser (background) with Supersonic ReCOIL Nozzle (in hand)

The advantage of lower operating temperature can be evaluated by observing that the threshold lasing condition is (17);

$$Y_{th} \frac{[O_2(^1\Delta)]}{[O_2(Total)]} = \frac{1}{2K_{eq} + 1}$$
 (18)

where K_{eq} is defined in Eq. 1b. At room temperature $Y_{th} = 0.15$, and in a Mach 2 flow $[T \approx 150 \text{ K}] Y_{th} \approx 0.04$, a considerable improvement in the power available [0.11 yield points].

Beam Quality improvements are less obvious. The energy in the laser is stored in the $O_2^{\ 1}\Delta$) which is nearly resonant with the upper laser level [$I^2P_{1/2}$, see Figure 1]. Since the ratio of iodine atoms to total oxygen is small [typically ≤ 0.05], each iodine atom is repumped many times throughout the flow field during the lasing process. Efficient power extraction requires large circulating fluxes resulting in short extraction distances [sugar scooping] and steep thermal density gradients which degrade beam quality. The higher velocities in supersonic COIL results in power extraction over longer stream wise distances and the circulating power and density variations will be more uniform across the optical aperture.

In 1980 when the initial supersonic COIL demonstration was being considered, sparger $O_2(^1\Delta)$ generators were the only well characterized sources to power the laser. Since higher operating pressures are required the transport volume would have to be minimized and smaller more efficient cold trap would have to be employed.

The most difficult issue, mixing the heavy secondary molecular I_2 into the primary $O_2(^1\Delta)$ stream, was addressed with a mach 2 nozzle where the I_2 is injected transverse to the primary stream in the subsonic region of the nozzle⁽⁴⁰⁾. The transverse subsonic injection enhanced mixing and helped initiate the auto-catalytic I_2 -dissociation mechanism proposed by Heidner. From a purely kinetic standpoint the dissociation should occur more rapidly in the high pressure subsonic section of the nozzle.

With the elements discussed above a 25 cm gain length device (ReCOIL) was designed, built, and tested at the Phillips Laboratory [see Figure 8]. Testing of this device resulted in the first successful demonstration of a supersonic COIL. A second sparger driven supersonic COIL was demonstrated in 1984 at TRW⁽³³⁾.

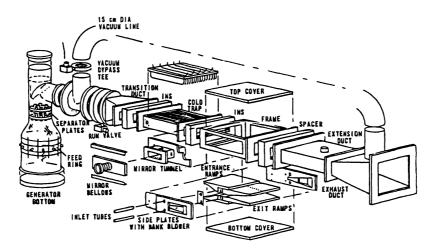


Figure 8. Schematic of the ReCOIL System

3.3 Phase III: COIL Engineering Demonstrations (1984-1989)

The essential features of the supersonic COIL are illustrated in Figure 9 and the four areas that required refinement are; (1) $O_2(^1\Delta)$ generator operation at high pressure, (2) efficient $O_2(^1\Delta)$ transport, (3) water removal, and (4) efficient $O_2^{-1}I_2$ mixing. These issues will be discussed in the following subsections.

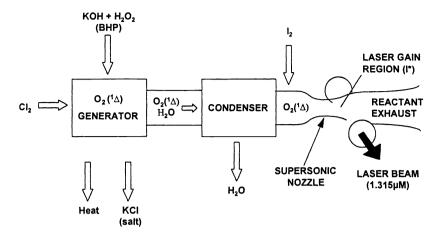


Figure 9. Supersonic Chemical Oxygen Iodine Laser

3.3.1 Rotating Disk $O_2(^1\Delta)$ Generators

The first supersonic COIL lasers were operated with sparger oxygen generators $^{(32, 33)}$, however improving the efficiency of the COIL using these generators is limited $^{(28)}$. An oxygen generator that has more BHP surface area, less generator volume, less transport duct volume, and more rapid BHP surface replenishment was needed. Although spray or aerosol reactors could potentially solve these problems, in the early 1980s, compact efficient spray $O_2(^1\Delta)$ generators had not been developed, and did not look particular promising $^{(41)}$. Harpole et. al. $^{(42)}$, developed a rotating disk oxygen generator [see Figure 10] where multiple, thin, disks were stacked together and partially immersed in a pool of BHP. The disks were rotated at 20 rpm wetting the disks with a BHP film [about 0.03 cm thick on each side]. The Harpole Roto Generator, described elsewhere $^{(42)}$, produced 40% to 60% $O_2(^1\Delta)$, 90% chlorine utilization with a chlorine flow rate of (0.5-0.6) mole/sec, He/Cl₂ = 3 or 4, and a generator pressure of 40 to 60 torr $^{(43-46)}$. This generator was an important development required to produce efficient supersonic lasers and an excellent review of its operation is reported by Dickerson et. al. $^{(47)}$ and Copeland et. al. $^{(48)}$.

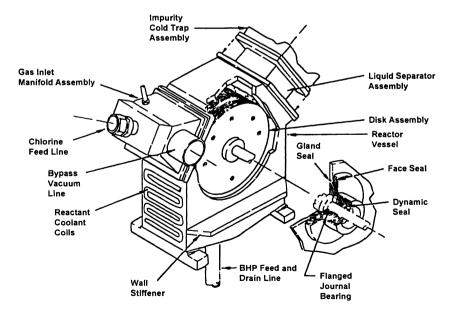


Figure 10. Harpole Rotating Disk Oxygen Generator (Ref. 42)

3.3.2 Water Vapor Control

The importance of water vapor control^(45, 46, 49) is shown in Figure 11 [note the monotonic power decrease as the water mole fraction is increased⁽⁵⁰⁾]. To minimize this effect two approaches have been used; (1) vapor cold traps to remove water and (2) lower BHP bath temperature to prevent water vapor formation. The cold trap method works well for subsonic lasers where the pressures are low (Torr range) and the added volume $[O_2(^1\Delta)$ residence time] between the oxygen generator and laser cavity do not contribute significantly to $O_2(^1\Delta)$ pooling [see Eq. 14a]. In supersonic COIL lasers, where the pressures are significantly higher (10's of Torr range), the pooling losses associated with any added transport volume become unacceptable⁽⁵¹⁾[note, in Eq. 15a, the quadratic dependence of pooling loss as $O_2(^1\Delta)$ pressure increases].

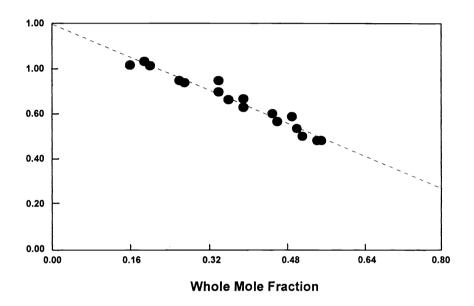


Figure 11. COIL Power as a Function of Water Mole Fraction (Ref. 50)

3.3.3 Supersonic O₂-I₂ Mixing Nozzles

There are four critical nozzle dimensions that require adjustment in a supersonic COIL nozzle; (1) the throat size and nozzle expansion ratio, (2) the I_2 injection hole size(s) and distribution, (3) the location of the I_2 injection hole(s) relative to the nozzle throat, and (4) the resonator location relative to the nozzle exit plane [see Figure 12]⁽⁴⁰⁾. The most surprising aspect of this design is its forgiving nature relative to more traditional HF/DF chemical lasers⁽⁵²⁾. A COIL nozzle with a 0.6 cm throat, an exit area to throat area ratio of 2:1, and a double set of sonic I_2 injection orifices located (1.0-1.3) cm upstream of the sonic throat works well over a wide range of operating conditions. In particular, nozzle plenum pressures as low as 11 torr (He/Cl₂ = 1)⁽³²⁾ and as high as 70 torr (He/Cl₂ = 4)⁽⁴⁶⁾ have been demonstrated. Power optimization with fixed nozzle hardware depends on $O_2(^1\Delta)$ generator performance, secondary flow rate and plenum pressure [I_2 penetration], I_2/O_2 ratio, flow composition, and water⁽⁵³⁾. However these nozzle designs have worked well when the parameter space is optimized⁽⁴⁶⁾.

Applying the LIF technique to supersonic nozzles shows how under penetrated secondary settings prevent the primary and secondary from ever fully mixing [Figure 13], and when the secondary fully penetrates to the flow center line before the nozzle throat, good I_2 mixing is evident at the nozzle exit plane⁽⁴⁰⁾.

3.3.4 The RotoCOIL Laser

The RotoCOIL laser [see Figure 14] represents the culmination of the engineering and efficiency demonstration phase of the COIL program. Three oxygen generators were used instead of one large one for reasons of expediency and in spite of such engineering short comings, the laser is the most efficient multiple kilowatt COIL ever built [Eff = $P_{\text{measured}}/(91 \text{ X } \dot{C} \text{ I}) = 0.25$] More detailed discussions of the performance of RotoCOIL are presented elsewhere (43-46, 54).

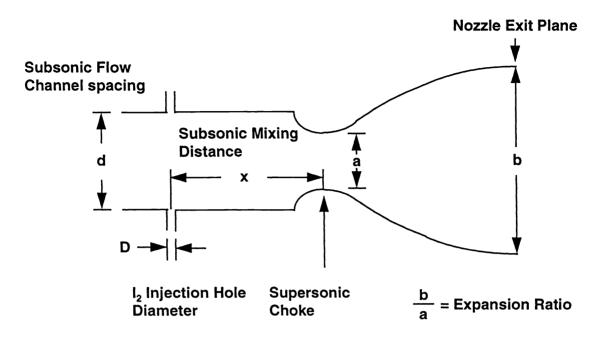


Figure 12. Critical Supersonic COIL Nozzle Dimesions (Ref. 40)

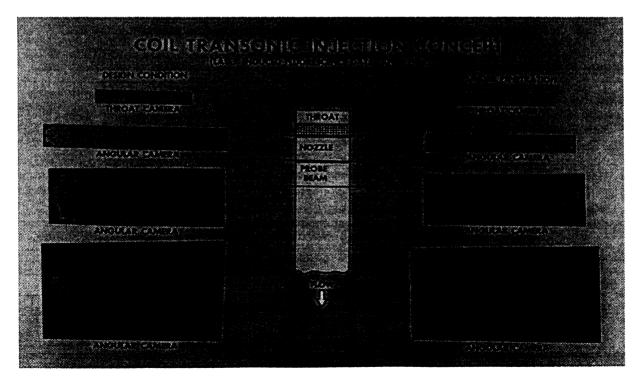


Figure 13. Supersonic COIL LIF Data (Ref. 69)

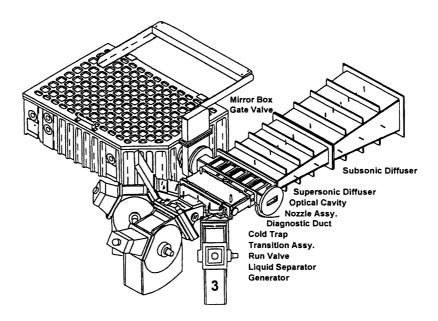


Figure 14. RotoCOIL Laser Device Isometric (Ref. 54)

The saturation and extraction behavior of the COIL laser requires a comment. The RotoCOIL laser saturation curve is shown in Figure 15 and although the Rigrod analysis can be applied^(45, 46, 54), the existence of a distributed loss in the gain medium is not experimentally well demonstrated. Mirror scattering and diffraction losses can also explain this saturation curve⁽⁵³⁾. This issue is still being investigated.

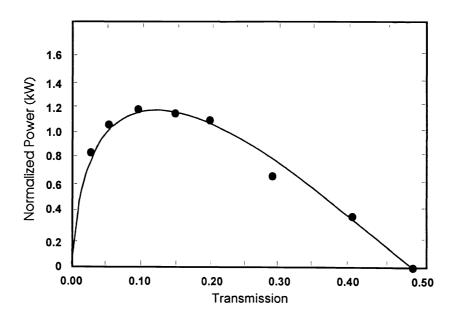


Figure 15. RotoCOIL Power vs. Transmission Curve (Ref. 45)

3.4 Phase IV: COIL Efficiency Improvements (1990-Present)

By the 1990s the focus of device development shifted from engineering demonstrations to device efficiency improvements. These improvements required a multifaceted approach, including modifying hardware, modeling, and developing new diagnostics. Modification of the COIL hardware has concentrated on the $O_2(^1\Delta)$ generators. The first area of generator improvement is thermal and salt management of the BHP solution. As mentioned earlier COIL performance is limited to short run durations [a few seconds] caused by the heat release in the oxygen generators and the subsequent water vapor build up (see Eq. 8 and Figure 11). To counter this effect cold BHP was flowed through the rotogenerator to control the temperature of the BHP reaction zone⁽⁵¹⁾. Steady power performance for up to four minuets was demonstrated using this methodology. Current closed-loop experiments have shown that recondition BHP in "real time" is possible [see Figure 16]⁽⁵⁵⁾.

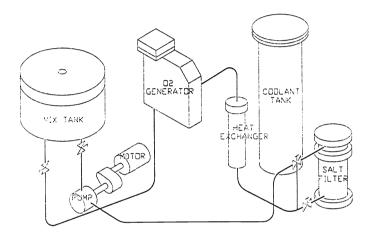


Figure 16. VertiCOIL Closed-Cycle BHP System Schematic

Further generator improvement requires methods to increase BHP surface area, reduce generator gas volume, and increase the reaction zone $[O_2H]$ replacement rate⁽²⁸⁾. During the 1980's spray reactors aimed at addressing these issues were postulated⁽⁵⁶⁾ but failed because the BHP acrosol could not be efficiently separated from the gas stream⁽⁴¹⁾. In 1988, development began on a new type of droplet reactor that was capable of producing droplets of uniform diameter, which greatly simplified the liquid separation process⁽⁵⁷⁾. Several versions of this generator have been tested^(58, 59) and by design the droplet generator is a flowing BHP system that will minimize H_2O production.

The early stages of COIL modeling in the USA concentrated on the gas phase [kinetics of $O_2(^1\Delta)$ reactions, I_2 dissociation, and cavity kinetics]. Excellent reviews of these studies are available $^{(29, 30, 34, 48, 60)}$. Although the three step Hurst mechanism (see Eqs. 5-7) for the Cl_2 -BHP reaction was proposed in 1978 and several investigators worked on measuring the reaction rates , it was not until recently that modeling emphasis turned to the Cl_2 /BHP diffusion/reaction mechanism and solving the coupled nonlinear differential equations describing the Cl_2 utilization and $O_2(^1\Delta)$ yield in terms of the Cl_2 and $O_2(^1\Delta)$ concentrations in the bulk gas and the O_2H^2 concentration at the surface of the liquid BHP (65, 65). Recent advances in computer memory and speed has also allowed tackling the 3-D Navier-Stokes analysis of the I_2 - O_2 mixing in supersonic COIL nozzles. For the first time an end to end analysis of COIL is at hand and early results are encouraging. An example of this progress is shown in Figure 17 which compares recent I_2 nozzle distribution predictions with I_2 LIF data taken 10 years ago⁽⁶⁹⁾.

Improving our fundamental understanding of COIL through improved diagnostic techniques continues to be an essential element of COIL development. Iodine dissociation⁽⁷⁰⁾, and small signal gain⁽⁷¹⁾ diagnostics have been developed and used on a slit nozzle configuration. In addition a new diode laser based water vapor diagnostic has been developed⁽⁷²⁾ and used on the same nozzle⁽⁷³⁾. A new absorption technique for accurately determining the yield of $O_2(^1\Delta)$ in the laser cavity is also being developed⁽⁷²⁾.

COIL work in the future will focus mainly on improving the chemical efficiency and BHP usage in order to reduce the size, weight, and costs of COIL. If COIL is to survive as a viable laser, transition to the private section will be an important objective during the next few years.

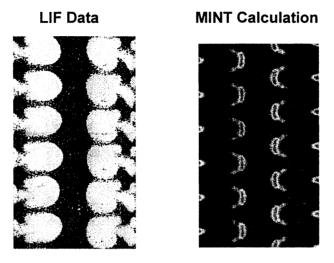


Figure 17. Comparison of I₂ Distribution in COIL Nozzles.

4. SPIN OFF COIL TECHNOLOGY

Two spin off COIL technologies will be discussed, frequency doubling and magnetic gain switching.

4.1 COIL Frequency Doubling

A series of extra cavity frequency doubling tests were performed using the RotoCOIL laser⁽²³⁾. The diffraction limited output from the IR laser was tightly focused into LiIO₃, [selected for doubling due to its large nonlinear optical coefficient, low absorption, and availability⁽⁷⁴⁾]. Crystal lengths of 1.1 cm and 2.2 cm were used and conversion efficiencies of 8% were achieved resulting in visible (657 nm) cw outputs of nearly 700 W [see Figure 18]. Catastrophic crystal failure occurred after a 1 sec exposure to a focused beam of 6.8 kW.

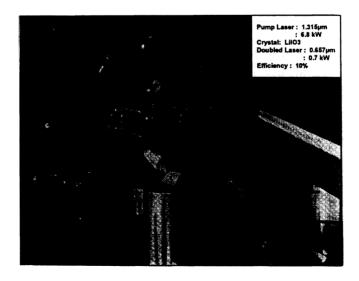


Figure 18. Photograph of the Red Laser Light at 657 nm from LiIo₃ Crystal Pumped With RotoCOIL 1.315 mm Laser.

4.2 Receptively Pulsed COIL

For the past several years the Phillips Laboratory has been developing a gain switched COIL (24, 25) and the field-nulling gain-switched concept is illustrated in Figure 19. The Figure shows theoretical calculations of the iodine hyperfine spectrum for zero field [Figure 19a], and for a 400 gauss magnetic field, P polarization [Figure 19b] and S polarization [Figure 19d]. Figure 19c shows schematically the hardware arrangement and the operating sequence is as follows. Initially a static magnetic field of 400 gauss is applied to the cavity by an external permanent magnet [Figure 19c]. The cavity out coupler is chosen so that the static magnetic field suppresses the gain below the lasing threshold condition. A fast rising current pulse is then applied to the field coils [Figure 19c] with a polarity that nulls out the cavity magnetic field. The gain suddenly rises above threshold to its zero field condition (Figure 20a) and a laser pulse is extracted from the medium. Once the laser pulse has been extracted, the current in the field coils is shut off turning the cavity field back on. The cavity refills with fresh gain media and the process is repeated producing a train of pulses.

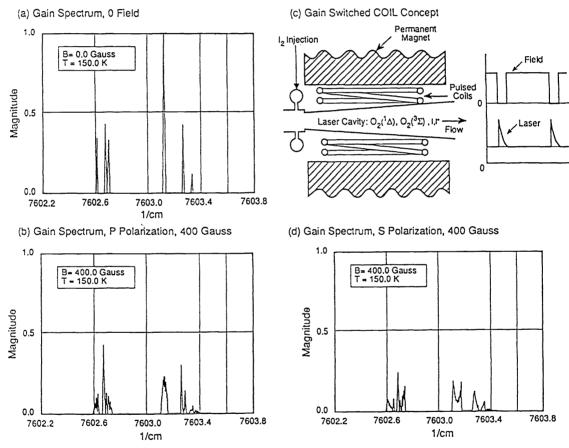
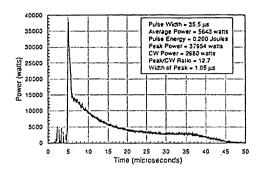


Figure 19. Concept of Nulling Gain Switch COIL

Figure 20 shows a sample data set for a 500 Hz gain-switch experiment. Figure 20a shows the temporal profile of a single laser pulse and Figure 20b shows the associated Helmholtz coil current [along with the estimated magnetic field strength]. In Figure 20, once the pulsed field has canceled the permanent magnetic field [approximately $(0.3 - 0.5) \mu s$] there is a time delay of about 3 μs before the power spike occurs; this is the cavity mode buildup time. The peak power $(W_{1/2} = 1 \mu s)$ is nearly 39 kW and represents the energy stored in the $I(^2P_{1/2})$. At the end of the gain switch spike, singlet delta $I(^2P_{1/2})$ repumping by $O_2(^1\Delta)$ and the cavity resonator parameters control the remainder of the pulse until the steady lasing begins at approximately 20 μs [Pcw = 3 kW]. At about 35 μs , the current pulse ramps down forcing the gain below threshold, shutting the laser off. The peak power enhancement [peak power/cw power] is about 13 and the integrated energy is 0.2 joules.

(a) Experimental Power Trace



(b) Experimental Field Coil Trace

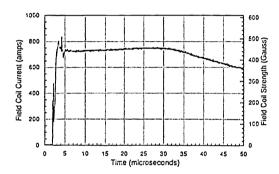


Figure 20. Experimentally Measured a) Laser Pulse, and b) Field Nulling Current Pulse.

5. CONCLUSION

In this paper we have reviewed the key technical developments leading to the invention and refinement of the COIL laser in the United States. The story covers the 34 year period between 1960 and the present. The current oxygen generator and nozzle concepts are proven and the laser best operates at the kW and higher level. The excellent fiber transmission makes the laser a candidate to be used in a situation where one (10-50) kW unit can feed several work stations⁽⁷⁵⁾. The laser operates at a good wavelength [1.315 μ m], offers excellent beam quality, and good beam deliverability [optical fiber transmission]. These characteristics along with the inexpensive chemicals that power the laser make COIL a viable candidate for industrial development.

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