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History of COIL development in Japan: 1982-2002

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ABSTRACT

A twenty-years of COIL researches and developments in Japan are reviewed. The researches of four major sites, namely, Keio University, Industrial Research Institute, Kawasaki Heavy Industries and Tokai University are presented in order of time. Epoch-making works are highlighted, and the significance of those works in the industrial COIL development is discussed. Finally, current status of COIL researches in Japan is introduced.

Keywords: iodine laser, COIL, Japan, history

1. INTRODUCTION

Chemical oxygen-iodine laser (COIL) is a continuous-wave chemical laser operating at 1.3µm. The principle of COIL is briefly described as follows. Hydrogen peroxide is mixed with some alkali (normally KOH or NaOH) and prepared in a vessel. The solution is called a BHP (basic hydrogen peroxide). Cl₂ gas is provided in it to produce singlet oxygen O₂(1Δ) by the following reaction,

\[ \text{H}_2\text{O}_2 + 2\text{MOH} + \text{Cl}_2 \rightarrow 2\text{H}_2\text{O} + 2\text{KCl} + \text{O}_2(1\Delta). \] (1)

Water vapor is released from BHP together with O₂(1Δ) and should be removed since it is a strong quencher of the lasing species, I(2P_{1/2}). Singlet oxygen is then mixed with iodine molecule vapor. Iodine is dissociated by the collisional energy transfer from O₂(1Δ). The iodine atom is then excited to the first electronically excited state by the subsequent collision with O₂(1Δ), and lasing is obtained between (2P_{1/2}) - (2P_{3/2}) levels of iodine. The I₂/O₂ ratio of typical COIL is a few percent and an iodine atom is excited many times in the laser cavity. The reactions are summarized as follows,

\[ n\text{O}_2(1\Delta) + I \rightarrow n\text{O}_2(3\Sigma) + 2I(2P_{3/2}) \] (2)
\[ \text{O}_2(1\Delta) + I(2P_{3/2}) \leftrightarrow \text{O}_2(3\Sigma) + I(2P_{1/2}) \] (3)
\[ I(2P_{1/2}) \rightarrow I(2P_{3/2}) + h\nu. \] (4)

Chemical efficiency is defined by the following formula,

\[ \eta_c = \frac{\text{[LaserOutput(W)]/h\nu}}{[\text{Cl}_2 \text{ mol flow(mol/s))]} N_A \times 100 \%}. \] (5)

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In principle, a Cl₂ molecule produces an O₂(1Δ) (eq.1) and that excites an iodine atom (eq. 3), then it produces a laser photon (eq. 4). Therefore, chemical efficiency is the quantum yield of the comprehensive chemical processes involved in COIL. It is very often used to measure the performance of a COIL device.

2. BEGINNING – EARLY 1980'S

The Faculty of Science and Technology, Department of Electric Engineering, Keio University has a long history of research in gas lasers that goes back to 1960’s. At the time when the laser laboratory was led by Fujioka, one of his interests was the photolytic iodine laser. Therefore, it was a natural consequence that he started the research of COIL shortly after the first demonstration of lasing by the United States Air Force.¹

Although the COIL was considered as a candidate of directive energy weapons (and is going to be realized soon), Fujioka was aware of its usefulness as an industrial laser from the very beginning of the COIL history. At the same time, Watanabe, who was a graduate student of Keio University Laser Laboratory, started his career at the National Defense Academy in Japan with the research of COIL, too. Japanese COIL history started with a race between a teacher and his student.

The student reached the finish line first. The first oscillation of COIL was obtained by Watanabe et al.² in 1982. Shortly after, Keio University also succeeded in lasing by Serizawa, Uchiyama and Fujioka et al.³ Figure 1 shows the singlet oxygen generator (SOG), water vapor trap, and optical cavity of the Watanabe’s apparatus. The constructions of the both COILs were so-called Bachar-Rosenwaks type,⁴ which was the typical design in the early 80’s. According to the Watanabe’s first report, the output power was 12W and chemical efficiency was 4.4%. Watanabe measured the property of the laser medium by the variable-loss optical cavity.⁵

Keio group’s interests were oriented to the industrial applications from the very beginning of their studies. It must be emphasized on that in the early 80’s, the use of COIL as an industrial laser was not widely accepted and Keio University pioneered the industrial COIL development. They have already demonstrated the optical fiber transmission of COIL output in 1983.⁶ To say the chemical efficiency, their early apparatus obtained 16.8% at an output of 105W from a 25×1cm² flow duct.⁷

3. KHI & IRI – LATE 1980’S

There was a remarkable movement in Japanese COIL history in 1986. Fujioka resigned Keio University and started-up a laser research center at Industrial Research Institute (the institute renamed itself as Institute of Research and Innovation in 1989). One of the objectives of the IRI Laser Laboratory was to offer the research site for national research programs of high-power industrial lasers. The well-known 20kW CO laser for decommissioning and dismantlement of nuclear power plants (D&D)⁸ was originated here. A five-year project of industrial COIL development also started up at the IRI Laser Laboratory, and the program was shared with Kawasaki Heavy Industries.
The goal of the project was the development of the world's first industrial COIL at 1kW output, all-day-long operation capability.

Initially, the research program was led by Yoshida. Yoshida modified the bubbler type SOG to improve the performance of the SOG that used dilute hydrogen peroxide. Although eq. (1) produces singlet oxygen at an efficiency of 100%, not all the oxygen is yielded in the $\left( ^1 \Delta \right)$ state because of the quenching reactions in the liquid phase. It was believed that dense (>60%) $\text{H}_2\text{O}_2$ should be used for the efficient yield of $\text{O}_2\left( ^1 \Delta \right)$.

The use of dilute hydrogen peroxide is crucial for industrial applications. In Japan, the hydrogen peroxide of equal to or less than 35wt% was much less restricted for usage, storage, and conveyance. Figure 2 shows the IRI's first COIL apparatus. Dense array of bubbler tubes close to the BHP surface is seen. Finally, Yoshida claimed (and not widely believed) that a 40% of chemical efficiency was obtained at an output power of 200W, using a 35wt% hydrogen peroxide.

The IRI COIL apparatus was scaled up to a 1kW device, then the know-how was transferred to KHI to make a commercial COIL device. The COIL research at IRI Laser Laboratory ceased in 1991.

4. KEIO UNIVERSITY – 1990'S

It can be said that Keio group rather stepped aside the main stream of the industrial COIL development in 1990's. Nevertheless, a number of unique works have been done during this decade.

In the early COIL devices, the unwanted byproduct of the SOG, water vapor, was removed by the cold trap (see Figs. 1 and 2). Uchiyama considered that the necessity of the water vapor trap inevitably prevents long-term operation of COIL, and should be removed. Uchiyama's strategy was the deep cooling of BHP (close to ~30°C), and use of a porous pipe SOG that was effective at very low BHP temperature. Figure 3 shows the schematic of the porous pipe SOG and COIL without a water vapor trap the porous pipe SOG and COIL without a water vapor trap. An output power of 87W with a chemical efficiency of 7.8% was obtained.

The unique porous-pipe SOG led Uchiyama to another application: pulsed high-pressure SOG. The principle of the pulsed SOG is as follows: first, the surface of the porous pipe is kept wet with BHP in a closed chamber. Then Cl$_2$ gas is injected to the inside of the porous pipe at a blast. The Cl$_2$ gas penetrates to the surface, react with the BHP at the surface of the pipe, and fill the chamber instantaneously with O$_2\left( ^1 \Delta \right)$. In this way, a 35Torr O$_2\left( ^1 \Delta \right)$ pressure was obtained.

Fig. 4: Pulsed COIL. 1: Cl$_2$ tank 2: Porous pipe 3: Perforated inner tube 4: Solution (H$_2$O$_2$+KOH) 5: Air filter 6: Iodione injectors 7: Manifold 8: Prechamber 9: Ge photodiode 10: Si photodiode 11: Pressure gauge
observed in a very short period.\textsuperscript{15} The world's first pulsed COIL was developed using this SOG.\textsuperscript{16} An ambitious study, observation of lasing from O\textsubscript{2}(^1\Delta) directly, was tried using this pulsed SOG.\textsuperscript{17} According to the theoretical calculation, a 6-m active volume length should produce a realistic positive gain even O\textsubscript{2}(^1\Delta) to O\textsubscript{2}(^3\Sigma) is a strictly prohibited transition. However, the experiment resulted in unsuccessful.

Q-switching\textsuperscript{18} and magnetic gain switching\textsuperscript{19} were studied at Keio University. Because COIL is an energy transfer laser, the lasing species has only a few percent of the extractable energy. Therefore, those switching methods do not work as efficient as say, CO\textsubscript{2} laser. Nevertheless, an 11 times enhancement in Q-switching regime, and a 3.3 times enhancement in magnetic gain switching were obtained respectively.

Intracavity second harmonic generation was studied at Keio University. The objective of this study was to get the more valuable visible light than infrared at an efficiency of 100%. The idea of the intracavity SHG is to place a SHG crystal inside the laser cavity and use dichroic mirrors (total reflector for fundamental and transparent to the second harmonic) for optical cavity. Because only the converted light can escape from the optical cavity, the SHG crystal acts as an output coupler. Therefore, 100% conversion efficiency is expected. The obtained output power was 16.1W in TEM\textsubscript{00} mode,\textsuperscript{20,21} and the conversion efficiency was estimated to be nearly 100%. It means that the second harmonic output was the same as the fundamental output in which an optimized partial reflector was used and the SHG crystal was removed.

Iodine predissociation was studied at Keio University. In the first step of the oxygen-iodine reactions (see eq. 2), approximately 10% of the O\textsubscript{2}(^1\Delta), that have the capability of pumping iodine, is used merely to dissociate iodine molecules to atoms. Therefore, substituting the dissociation with external energy enhances the output power of COIL because more O\textsubscript{2}(^1\Delta) survives to the laser cavity than conventional COIL. The point is that the theoretical limit of enhancement is not limited to 10% because there is an oscillation threshold of O\textsubscript{2}(^1\Delta) yield in COIL same as the other lasers. The extractable power \( P_{\text{ext}} \) from the singlet oxygen flow can be expressed as

\[
P_{\text{ext}} = \dot{m} E \eta_q (Y - Y_{\text{th}} - Y_{\text{dis}}),
\]

where \( \dot{m} \) is the oxygen flow rate [mol/s], \( E \) is the energy of singlet oxygen per unit quantity [J/mol], \( \eta_q \) is the quantum efficiency of the energy transfer (0.97), \( Y \) is the yield of the oxygen, \( Y_{\text{th}} \) is the lasing threshold yield, and \( Y_{\text{dis}} \) is the part of the singlet oxygen necessary for iodine dissociation. \( Y \) is 0.4 to 0.6 for the typical SOGs. We assume \( Y \) to be 0.5, \( Y_{\text{th}} \) to be 0.2 (for subsonic COIL), and \( Y_{\text{dis}} \) to be 0.1. In this case, the extractable power comes to \( P_{\text{ext}} = 0.2 \dot{m} E \eta_q \). If we completely substitute the dissociation energy with the external source, \( Y_{\text{dis}} \) is no longer subtracted and the extractable power becomes \( P_{\text{ext}} = 0.3 \dot{m} E \eta_q \). Therefore, approximately 50% of output power enhancement is expected by the external iodine dissociation.

Uchiyama used an RF discharge to dissociate iodine and applied the technique to their subsonic COIL. As a result, chemical efficiency was improved from 4% to 12%.\textsuperscript{22}

5. KHI - 1990'S

KHI started the development of industrial COIL with IRI in the late 80's.\textsuperscript{23,24} The national project delivered the world's first commercial COIL in 1992.\textsuperscript{25} The device was sold to the Applied Laser Engineering Center (ALEC), and it is still available for value. The remarkable capability of the device was the eight hours of long-term operation at a stable
(±2%) output power of 1kW. To cope with the water vapor trap clogging, KHI developed a breakthrough disk type water vapor trap as seen in Fig. 6. Because the ice deposited on the surface of the disks are continuously removed by the scraper, the cross section of the gas flow duct is kept constant. As far as we know, this device is still holding the continuous-operation time record.

After the successful end of the national project, KHI continued the development of COIL. It was the time that the U. S. Air Force demonstrated the supersonic COIL operation.\(^{26}\) The application of supersonic expansion has significant advantages for industrial COIL, too. Especially, high output power scalability and high output power per unit width of the flow duct leads to a realistic industrial device at an output range of >10kW. In addition, the requirement of the high-pressure SOG operation removed the necessity of the water vapor trap, because the water vapor pressure is limited to the saturation vapor pressure of water in spite of the increased operating pressure. However, the traditional bubbler type SOG could not operate at higher pressure than a few Torr. KHI have selected the application of transverse type jet SOG for their supersonic COIL. In 1996, they have obtained an output power of 10kW.\(^{27}\) and 12kW was recorded in 1999. This remarkable output was obtained with a very high efficiency, \(\eta=26\%\). Moreover, the output power was stable for more than 30 minutes.

It was not only the COIL development itself but also the applications of COIL that were pursued at KHI. Because COIL is a chemical laser, its application to the D&D is very promising. Since no high-power electricity is required for COIL and the nuclear power plants are usually settled in remote places, its advantage to D&D is obvious. In addition, the wavelength of 1.3μm, that is ideal for optical fiber delivery, is suitable for remote, unmanned cutting. Needless to say that the high-power capability and high beam quality of COIL is advantageous for cutting of thick steel or concrete. KHI conducted number of studies\(^{28-30}\) for material processing of COIL. They have proposed an empirical formula that predicts the cutting performance of very thick materials.\(^{31}\)

Recently, KHI demonstrated an underwater cutting of thick steel specimen by the COIL through optical fiber delivery.\(^{32}\) At an output power of 7kW, cutting capability for stainless steel workpiece was up to 80mm of thickness.

![Photo 1: KHI's supersonic COIL operating at 12kW.](Image)

![Fig. 6: KHI's subsonic COIL with a rotating-disk type water vapor trap.](Image)

![Fig. 7: Experimental setup of the COIL D&D - underwater cutting with optical fiber delivery of laser power.](Image)
The history of COIL research at Tokai University began in 1996, shortly after Fujioka moved to Tokai University and started up a laser research laboratory. Tokai University COIL research group took up a research program sponsored by NEDO (New Energy and Industrial Technology Development Organization) together with KHI. The goal of the program was to show the feasibility of the “Optical Power System” (See Fig. 8). The name implies the use of laser power like today’s electricity. High-efficiency operation of supersonic COIL, output power enhancement by using microwave discharge, output power control, optical fiber transmission, high beam quality resonator, and reuse of BHP were studied.

To start up COIL research at Tokai University, Zagidullin was invited and their counter-flow jet type SOG technology was transferred. It was soon after the start-up of the project that the very high chemical efficiency (23.4%) of the supersonic COIL using nitrogen as buffer gas was obtained. At that time, supersonic COILs were operated using helium as buffer gas to obtain supersonic expansion. The result showed that inexpensive nitrogen could be also used for supersonic COIL and that could dramatically reduce the operation cost. Figure 9 shows the first version of Tokai University’s COIL apparatus.

Sugimoto demonstrated the output power stabilization and modulation of COIL for the first time using Zeeman effect. Figure 10 shows the schematic. The laser power is monitored by a photodetector, whose output signal is compared to a reference signal to produce an acting error signal. The actuating error signal is then fed into the control unit of the variable dc power supply connected to the electromagnet, to change the magnetic field intensity. In this manner, the feedback system attempts to reduce the acting error signal in order for the output power to follow the reference signal.

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Fig. 8: Schematic drawing of the “Optical Power System”.

Fig. 9: SOG, iodine injector and supersonic nozzle of the Tokai University’s first COIL.

Fig. 10: Output power stabilizing system of COIL.

Fig. 11: Output power stabilization of COIL. Top: Fluctuated output by the added vibration. Bottom: Stabilized output.
Therefore, one can stabilize or actively modulate the output power to an arbitrary waveform. Figure 11 shows the typical result. An artificially generated output power fluctuation was stabilized within 0.4%. COIL output was modulated to an triangular waveform at 20Hz. Wani demonstrated the operation of COIL in a high-pressure subsonic regime. Although the supersonic operation was the major breakthrough of the COIL, the requirement of larger vacuum pump than subsonic regime is undesirable for many applications. Especially, the major advantage of COIL over electrically excited lasers, the mobility may be deteriorated by the large vacuum pump. The motivation of the high-pressure subsonic operation was the drastic reduction of vacuum pump by means of increasing the operation pressure of COIL and reducing the amount of buffer gas that was necessary to operate COIL in supersonic regime. The high-pressure operation capability of jet SOG was helpful for this operation regime. The main advantage of high-pressure subsonic regime to the good old subsonic COIL is device size and water vapor trap requirement. Thanks to the high-pressure operation of SOG, no water vapor trap was required upstream the laser cavity. The area of the flow duct per output power was the same as the supersonic COIL because the high pressure compensated the slow flow velocity. In fact, the subsonic COIL that used the same flow duct as the supersonic COIL obtained a higher output. The pressure at the laser cavity was 6Torr, and the chemical efficiency reached to 25%. Finally, the subsonic operation at 12Torr using a larger device was demonstrated.

In 1997, Tokai University and VNIIEF (Russian Federal Nuclear Center) conducted a joint research of a unique SOG, twisted aerosol SOG (TA-SOG). That was a part of ISTC (International Science and Technology Center) program. It was demonstrated that the TA-SOG had the potential to obtain much higher operating pressure than jet SOG without BHP mist entrainment. Now VNIIEV group demonstrated a quite high-efficiency (~30%) operation of COIL using TA-SOG.

The predissociation of the iodine molecule was studied at Tokai University, too. In the early research Endo pointed out theoretically that the supersonic COIL with subsonic mixing regime is unsuitable for the predissociation of iodine, because the dissociated iodine is a very fast quencher of O2(1Δ) and iodine must be in molecular state before the expansion. Soon after that, Endo demonstrated the transonic mixing of dissociated iodine, and supersonic injection of dissociated iodine to reduce the mixing point pressure, and enjoy the effect of the predissociation in the supersonic COIL. However, another problem arose. The complete mixing of primary flow (oxygen) and secondary flow (iodine) is much more difficult in the supersonic mixing regime. Tokai University is trying an elaborated mixing scheme proposed by Masuda, however, not yet successful to obtain as good output power as conventional supersonic COIL.

Possible applications of COIL were discussed at Tokai University. Sugimoto conducted the drilling of some nonmetallic materials (concrete, wood, rock, etc...) to assess the capability of COIL for the use of civil engineering and rescue for the disaster. Tei discussed the conceptual design of a D&D COIL based on the current COIL technology.

7. DEVELOPMENT OF SOG

Singlet oxygen generator has been extensively studied from the very early days of COIL research in Japan. The very high efficiency operation of COIL with dilute BHP owed to the Yoshida's modified bubbler SOG. Yoshida also studied an aerosol type SOG. Although he obtained a good singlet oxygen yield (80%), the problem of the aerosol separation from the gas flow was left unsolved.
Takehisa (Keio Univ.) developed the unique porous pipe SOG. The successful operation of COIL without a water vapor trap greatly owed to the porous pipe SOG. The porous pipe SOG technology was applied to the realization of the pulsed SOG, which generated a 35 Torr of singlet oxygen. Currently, Keio University group is studying some jet SOG variation. One of the ideas was the jet SOG with guide plates.

KHI’s COIL development started with the modified bubbler SOG that was developed in IRI, and the world’s first commercial COIL utilized it. Soon after that they developed the transverse-flow jet SOG for supersonic COIL. The property of their transverse jet SOG is the extraordinary stability; it can operate stably for more than an hour. It should be noted that the long-term operation of jet SOG is still an up-to-date topic of COIL development. Tokai University started COIL researches with the jet SOG that was transferred by Nikolaev and Zagidullin in 1997. Since then, Tokai University’s jet SOGs are following Zagidullin’s design criteria. In 1997, VNIEF’s original SOG, TA-SOG was tested at Tokai University, and its high performance was demonstrated. Now Tokai University is developing a new type of SOG, mist SOG. (Fig. 13). The goal of the mist SOG development is the stoichiometric usage of BHP at a single pass of the reaction zone. The unique counter-flow concept is the key technology to separate the BHP mist from the gas flow.

Fujii’s group at Anan National College of Technology is studying a completely different SOG, discharge type SOG. Now the yield of the singlet oxygen is very close to the oscillation threshold. Table I shows the timetable of SOG development in Japan.

Table I: SOG developments in Japan.

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8. THEORETICAL WORKS

Numerical simulation of COIL was mainly conducted at the Nagaoka University of Technology. A three-dimensional, Navier-Stokes code including chemical reactions was developed by Masuda. The problem of supersonic COIL attracted him because it is a unique problem from the viewpoint of fluid dynamics, since the Reynolds Number is much lower than the well-studied supersonic flows. Their studies began with the diagnostics of the duct flow, and followed by the...
discussion of the chemical reactions at the mixing nozzles. The results were fed back to the KHI-COIL. The role of the water vapor condensation and heat release to the laser medium property was theoretically discussed. The optical resonator was modeled using a one-dimensional wave optics, and the result was compared with a geometric optics model. The Q-switch operation of COIL was discussed from the viewpoint of fluid dynamics.

The possibility of the nuclear fusion driver using pulsed COIL was theoretically discussed by Endo and he predicted a new energy extraction regime, "quasi-cw energy extraction" with a 10ns pulse in the very high-pressure oxygen-iodine laser medium.

Endo discussed the possibility of the direct lasing from \( \text{O}_2(1\Delta) \). The point-source model included the estimation of the transition probability from each rotational level based on the H"on-London factor, time-dependent \( \text{O}_2(1\Delta) \) number density of each rotational level, and pressure dependent line broadening. According to the calculations, the highest gain was obtained at the \( ^0\text{Q}(9) \) transition (9→9,9), and the expected small signal gain from a 30Torr SOG was \( 3\times10^{-6}\text{cm}^{-1} \).

At Tokai University, Endo developed a numerical code that did not include the numerical fluid dynamics. The rather simple code was however very helpful to assess the behavior of the laser medium of the iodine predissociation, magnetic gain switching, and high-pressure subsonic operation.

9. NOW AND FUTURE – 2000–

The fruits of NEDO project conducted by Tokai University and KHI are now taken over to a private company, Miki-pulley. The company is known for the power transmission equipments, electric crutches, brakes, and so on. Their demand for a new product in their new business plan matched Fujioka's passion of industrial COIL development, and a five-year research project funded by JST (Japan Science and Technology Corporation) had started from FY2000. The goal of the program is the development of the first commercial supersonic COIL whose output is 5kW. A prototype is very close to the first light at the end of 2001. Tokai University participated in the program as a technical advisor.

In the last few years, KHI concentrated their studies on the applications of COIL. One of their research programs is a part of the national project, "Photon Project". High-speed welding of very thick aluminum alloy was demonstrated. The results of the KHI's latest work is presented elsewhere.

10. SUMMARY

A twenty-years of researches and developments of COIL in Japan were reviewed. The history of COIL in Japan originated at Fujioka's laboratory in Keio University. The first oscillation was recorded at National Defense Academy in Japan, and Keio University obtained lasing shortly after that. In 1986, IRI (Industrial Research Institute) and KHI (Kawasaki Heavy Industries) received a national project for development of the world's first commercial COIL. Now the product is working at ALEC (Applied Laser Engineering Center). Through the 90's, Keio University conducted number of unique COIL studies such as COIL without water vapor trap, porous pipe SOG, Q-switching, magnetic gain switching, intracavity second harmonic generation, pulsed operation, and so on. Tokai University COIL group started in 1996, founded by Fujioka. They obtained a high-efficiency (23.4%) operation of supersonic COIL with nitrogen as buffer gas, high-pressure subsonic operation, output power stabilization, and output power enhancement by the predissociation of iodine. Now the hottest COIL research program is conducted at Miki-Pulley. The goal of the project is the development of a commercial supersonic COIL at 5kW output.

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