

LASERS

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Pulsed chemical oxygen – iodine laser initiated by a transverse electric discharge

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***Abstract*. A pulsed chemical oxygen – iodine laser with a volume production of atomic iodine in a pulsed transverse electric discharge is studied. An increase in the partial oxygen pressure was shown to increase the pulse energy with retention of the pulse duration. At the same time, an increase in the iodide pressure and the discharge energy shortens the pulse duration. Pulses with a duration of 6.5 μs were obtained, which corresponds to a concentration of iodine atoms of 1.8×10^{15} cm^{-3}. This concentration is close to the maximum concentration attained in studies of both cw and pulsed oxygen-iodine lasers. A specific energy output of 0.9 J litre^{-1} and a specific power of 75 kW litre^{-1} were obtained. The ways of increasing these parameters were indicated. It was found that SF_6 is an efficient buffer gas favouring improvements in the energy pulse parameters.**

***Keywords*:** *chemical oxygen – iodine laser, transverse electric discharge, singlet oxygen.*

1. Introduction

At present, a chemical oxygen – iodine laser (COIL) is considered as a potential candidate for industrial applications. Due to a high output power at a wavelength of 1.315 μm, which corresponds to low-loss quartz optical fibres, this laser holds promise in situations where the radiation is to delivered to hazardous and not easily accessible areas, e.g., for the dismantlement of obsolete nuclear reactors [1].

By an industrial COIL laser is usually meant a laser operating in the cw mode. The capacity to operate in the pulsed mode would expand the area of application of the laser, especially involving processes where the power is the determining parameter (cutting materials, drilling, nonlinear frequency conversion, etc.). While on the subject of the pulsed mode, we will bear in mind the achievement of the maximum possible output pulse power. To do this, one should create an active medium with the maximum possible energy storage and then extract the stored energy in a minimal time.

There exist two approaches for obtaining the pulsed mode of operation of an COIL. The first approach involves different ways of amplification (loss) modulation or mode locking in a cw COIL. In this case, an active medium is used, which is obtained by mixing singlet oxygen with molecular iodine. The presence of fast relaxation processes prevents the formation of a large-volume active medium. One can easily show [2] that in this case the ratio between the pulse power W_p and the cw output power W_{cw} has the limit:

$$\frac{W_p}{W_{cw}} = \frac{K_f[I]L}{v}, \tag{1}$$

where K_f is the rate constant of the energy transfer from excited oxygen molecules $O_2(^1\Delta)$ to iodine atoms; [I] is the concentration of iodine atoms; L is the resonator length along the flow direction; v is the flow velocity. The limit achieved experimentally is equal to ~ 16 [3].

The second approach implies the formation of a large-volume active medium using a relatively stable mixture at the stage of filling the working volume. This can be accomplished by replacing a molecular iodine with an iodide, which does not dissociate upon mixing with singlet oxygen. The iodine atoms required for the operation of an COIL are produced upon pulsed decomposition of the iodide with the release of iodine atoms. To accomplish this, one can employ photolysis [4], an electric discharge [5], radiolysis, or an electron beam. This technique of the volume production of iodine atoms allows the formation of an active medium ~ 1 m in length with an oxygen pressure of several torr.

The iodine atoms produced in this way convert the energy stored in the active medium into the output laser energy in a time

$$\tau_p \simeq \frac{1}{K_f[I]}. \tag{2}$$

The output laser energy is determined only by the energy stored in the active medium and remains constant for the same singlet-oxygen concentration, whereas the period of energy extraction is inversely proportional to the concentration of iodine atoms produced. Therefore, by increasing the atomic iodine concentration, it is possible to augment the output pulse power (and the gain).

The use of photolysis for producing iodine atoms from the alkyliodide CH_3I resulted in a specific energy output of 3.3 J litre^{-1} for an oxygen pressure of 3 Torr and a content of singlet oxygen of 40 %. The reduced specific output energy corresponds to an energy extraction efficiency of 90 % and a chemical laser efficiency of 23 %. A chemical efficiency of 36 % was achieved for a lower oxygen pressure and, hence, for a higher content of singlet oxygen. The high chemical efficiency is due, in particular, to the absence of the

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chemical dissociation of iodine molecules and singlet-oxygen losses related to it.

The duration of a laser pulse could be varied from 15 to 500 μs for a virtually invariable pulse energy. A pulse output power of 300 kW was obtained with an active volume 1.4 litre. Therefore, the feasibility of producing an active medium with a length of ~ 1 m in the flow direction for an oxygen pressure up to 3 Torr was demonstrated, which made it possible to obtain a pulse power exceeding by three orders of magnitude the cw output power for the same consumption of the reagents.

Photolysis is a very convenient tool for producing iodine atoms because of the high selectivity of its action. However, it is not devoid of disadvantages. The primary disadvantages are the low pulse repetition rate in the repetitively pulsed mode and the low efficiency. An electric discharge provides the operation at pulse repetition rates in the kilohertz range. However, *ab initio* it was not clear whether the electric discharge can generate iodine atoms without an appreciable quenching of singlet oxygen. Our experiments proved that a longitudinal electric discharge can be efficiently used for this purpose. A specific energy of 1 J litre^{-1} was obtained for an oxygen pressure of 2 Torr. The electric efficiency of the laser (the ratio of the output laser energy to the electric energy stored in the capacitor) was 91 % [6]. We demonstrated the COIL operation in the repetitively pulsed mode with the repetition rate up to 20 Hz, the pulse repetition rate being limited by the power source.

The specific energy achieved using the discharge is lower than in the case of photolysis. This can be explained by quenching of singlet oxygen by electrons and the dissociation fragments of the mixture components or by heating of the active medium due to a high energy input required to produce iodine atoms. The energy of a UV photon that produces the dissociation of iodide is 5 eV. By assuming that all the energy stored in the capacitors is deposited in the active medium, one can estimate the energy 'cost' of one iodine atom obtained in the discharge. Under our experimental conditions, this 'cost' amounted to 25 eV.

The successful operation of the pulsed COIL with a volume production of iodine atoms in a longitudinal electric discharge stimulated the study of the feasibility of employing a transverse electric discharge for this purpose. A discharge of this type permits an operation at high oxygen pressures provided by contemporary singlet oxygen generators (SOGs), e.g., of the jet type, which gives grounds to expect an increase in the specific energy output. However, the use of a transverse discharge in an COIL operating at a relatively low pressure involves difficulties related to matching the discharge to the pulsed power supply.

2. Experimental

Different schemes for producing a transverse discharge are employed to initiate high-pressure gas lasers (CO_2 and excimer lasers). An electron-beam or UV preionisation is used in almost all of the schemes.

Considerable recent progress has been made in the initiation of a non-chain HF laser by a transverse discharge using an array of electrodes with a strong edge nonuniformity in the absence of preionisation [7]. A uniform volume discharge was obtained in a mixture of composition $SF_6 : C_2H_6 = 20 : 1$ for a total pressure of 160 Torr. The active volume of the discharge chamber was 50 litre for an electrode separation of 27 cm. The formation mechanism of a uniform volume discharge proposed by the authors of Ref. [7] gave promise that similar results may also be obtained in the case of a pulsed COIL. Like in an HF laser, the active medium of a pulsed COIL contains, in fact, an electronegative component – oxygen – and an easily ionisable (and electronegative) component – RI. We note that the ionisation potentials of CH_3I (9.5 eV) and CF_3I (10.2 eV) are lower than that of C_2H_6. Moreover, the low operating pressure of an COIL appeared to be better suited to the attainment of a uniform discharge.

Unfortunately, all our attempts to obtain a uniform discharge in the active medium of an COIL using the above-described electrode system have not met with success. Despite this fact, we believe that this approach holds promise in going over to higher pressures and longer electrode separations.

All the laser experiments were conducted using a pin cathode with a resistive discharge stabilisation. This discharge chamber was previously employed to initiate a non-chain HF laser. An assembly of 120 resistors (2.5 kΩ each) of the TVO-2 type was used as the cathode. The length of the cathode was 19 cm, the electrode separation was 1.8 cm, and the active volume of the laser cell was 52 cm^3. A voltage up to 18 kV was applied to the discharge gap from capacitors with capacities ranging from 3.4 to 20.4 nF via a TGI1-16/500 thyratron.

The singlet oxygen was produced in a chemical generator of the sparger type, which was a 230-mm long quartz cylinder with an internal diameter of 140 mm. A molecular chlorine was conducted from a soft bag, made of a polyethylene film, to the generator through its perforated bottom. The use of this bag allowed the pressure at the inlet to the gas system to be maintained equal to the atmospheric one with a high precision, thereby stabilising the chlorine flow rate.

To intensify the mass transfer, the generator was filled with a packing of Teflon Raschig rings measuring 12 mm × 12 mm × 1.5 mm. The packing measured 130 mm in height. Typically, the working composition was prepared by mixing 750 mlitre of a 50% hydrogen peroxide and 400 mlitre of a 50 % aqueous solution of KOH. The temperature of the working solution was initially −20°C and was raised during runs. The runs usually lasted from 10 to 20 s. After every run, the working solution was cooled down to the initial temperature.

The CH_3I iodide was delivered from a stainless steel vessel with a volume of 8 litre into which 100 mlitre of a liquid alkyliodide was poured. Perfluoroalkyliodide CF_3I, which has a high saturation vapour pressure, was delivered directly from a vessel. The He, N_2, Ar, and SF_6 buffer gases were admixed to chlorine upstream of the SOG. This allowed us to lower the partial chlorine pressure in the SOG and thereby raise the content of singlet oxygen in the gas flow.

The gas flow from the SOG was transported to the laser cell through a quartz tube 50 mm in diameter. The pressure gauge, the injector of iodide, and the inlet opening of the laser cell were separated from the SOG by distances of 500, 640, and 800 mm, respectively. The gas flow entered the medium segment of the T-shaped optical cell and was evacuated through its ends, at which the mirror mount units were located. The total length of the optical cell was 1600 mm and was equal to the resonator length with the use of internal mirrors. The discharge chamber was placed in one of the

arms of the optical cell previously employed in experiments with a pulsed COIL in the longitudinal geometry [6]. The time characteristics of lasing were recorded with the aid of a Ge photodiode with an integrating sphere, the output energy was measured with an IMO-2N calorimeter.

3. Experimental results

We studied the influence of the experimental conditions on the energy characteristics of the laser: the discharge energy, the applied voltage, the oxygen pressure, the sort and the pressure of the buffer gas. All experiments were performed with a resonator formed by a totally reflecting mirror and a mirror with a transmittance of 4.5 %. This value of transmittance was chosen on the basis of a preliminary investigation undertaken to optimise the resonator.

As follows from the principle of COIL operation, the singlet oxygen is the source of laser energy, and therefore the oxygen pressure in the active medium and the content of singlet oxygen determine the output laser energy. Fig. 1 shows the oscilloscope traces of laser pulses for different oxygen pressures. One can see that the time characteristics of lasing – the pulse duration and the time delay of the onset of lasing – are independent of the oxygen pressure. This implies that the concentration of discharge-produced iodine atoms remains nearly constant.

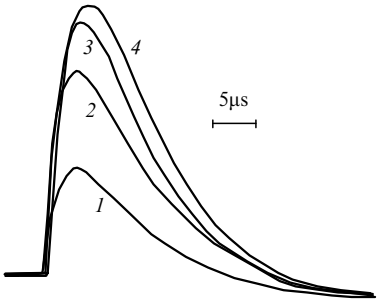

Figure 1. Output COIL pulses for a pump energy of 2.2 J and pressures $p_{N_2} = 9$ Torr, $p_{CH_3I} = 0.5$ Torr, $p_{O_2} = 0.5$ (*1*), 1.0 (*2*), 1.6 (*3*), and 2.0 Torr (*4*).

Therefore, the energy deposition into the laser active medium is determined primarily by the buffer gas whose partial pressure significantly exceeds the oxygen pressure. Unlike the pulse duration, the output laser energy is a function of the oxygen pressure (Fig. 2). A numerical model of this laser predicts, at least for the pressure range studied, a linear dependence of the output energy on the oxygen pressure for a constant content of singlet oxygen. However, as is seen from Fig. 2, the luminescence intensity does not grow linearly with pressure above 1.5–2.0 Torr, which testifies to a reduction of the content of singlet oxygen and explains the departure of E_{out} from the linear dependence. This behaviour is typical of a sparger SOG, which exhibits a strong pressure dependence of the content of singlet oxygen even in the low-pressure range ($\sim$ 1 Torr). A significant rise in specific energy output would be expected in going over to jet SOGs in which the effect of the operating pressure on the content of singlet oxygen is not that strong [8].

From relationship (2) it follows that shortening the laser pulse duration calls for a rise in atomic iodine concentration

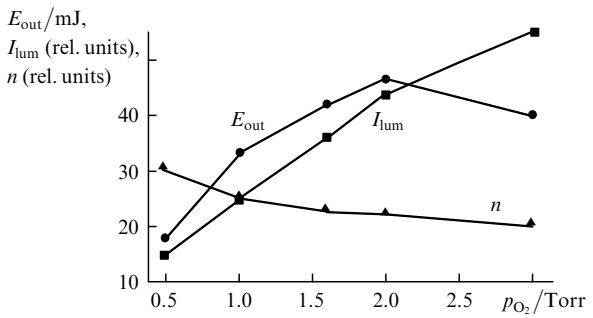

Figure 2. Output energy E_{out}, intensity of the luminescence of singlet oxygen I_{lum}, and the content of singlet oxygen n as functions of the oxygen pressure p_{O_2} for a pump energy of 2.2 J, $p_{N_2} = 9$ Torr, and $p_{CH_3I} = 0.5$ Torr.

in the active medium. This can be accomplished both by rising the iodide concentration for an invariable degree of dissociation and by increasing the degree of dissociation, i.e., by a growth of the energy deposition in the active medium. We note that in the former case there emerge complications associated with the provision of a high partial iodide pressure caused by the low saturated vapour pressure of typical alkyliodides.

We studied the CF_3I and CH_3I iodides. Fig. 3 shows the dependences of the pulse duration and the time delay of the laser pulse relative to the initiating pulse on the CF_3I pressure for the above iodides. Attention is primarily drawn to the fact that the efficiencies of both iodides as donors of atomic iodine are nearly equal, i.e., the iodine atom concentration produced by the discharge is independent of the sort of iodide. This leads to the coincidence of the time characteristics of the laser pulses. However, the energy characteristics of the laser pulses obtained with the use of different iodides are significantly different. In particular, under similar experimental conditions, the output energy provided by CH_3I was nearly 25–40 % higher than by CF_3I. As shown in Ref. 9, the reason for this difference lies with the quenching of singlet oxygen by RO_2 radicals ($R = CH_3$, CF_3) produced upon the iodide dissociation.

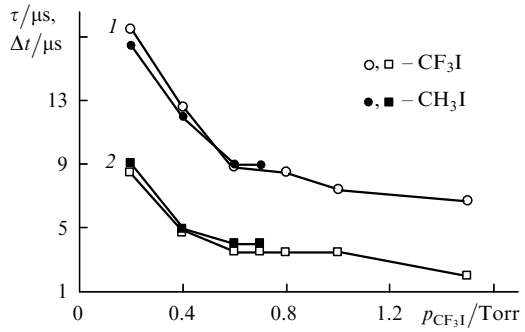

Figure 3. Dependences of the pulse duration τ (*1*) and time delay of the laser pulse Δt (*2*) on the pressure for different iodides CF_3I and CH_3I and a pump energy 2.2 J, $p_{N_2} = 9$ Torr, and $p_{O_2} = 1.0$ Torr.

Relationship (2) allows one to estimate the discharge-produced iodine atom concentration and, consequently, to determine the degree of dissociation of the iodide. The analysis performed shows that the degree of dissociation

lowers with increasing the iodide pressure – from 12 % for 0.2 Torr to 4 % for 1.5 Torr (the discharge energy is 2.2 J, the oxygen pressure 1.0 Torr, and the nitrogen pressure 9 Torr). We note that the iodine atom concentration corresponding to the minimal duration of the laser pulse equal to 6.5 μs amounts, by relationship (2), to 18×10^{15} cm^{-3}, which is close to the maximum concentration attained in COILs.

As noted above, the concentration of iodine atoms produced in the active medium depends on the energy deposited. In fact, increasing the energy stored in the power supply up to 3.3 J permitted the pulse duration to be shortened to 6.5 μs. The discharge energy was changed by varying the charging voltage as well as the capacitance of the power supply. No significant effect of these parameters on the output laser energy was observed in the voltage (10 – 18 kV) and capacitance (3.4 – 20.4 nF) ranges investigated. The pulse energy hardly changes when the threshold is sufficiently exceeded.

Unlike a photolysis-initiated laser where the role of a buffer gas reduces to increasing the heat capacity of the working medium and, hence, to lowering the operating temperature, which is of importance for the operation of a COIL, the role of a buffer gas in the case of discharge initiation is substantially more complicated. Apart from the influence on the heat capacity of the active medium, the presence of the buffer gas changes the plasma parameters, the resistance of the discharge gap, and hence the matching of the discharge to the power supply. In fact, raising the pressure of the active medium increases the resistance of the discharge gap and thereby increases the energy deposition, resulting in a reduction of the pulse duration, i.e., an increase of the concentration of iodine atoms.

The performance of a buffer gas from the standpoint of energetics depends on its sort. Fig. 4 shows the dependences of the output laser energy on the partial pressure of a buffer gas for a pump energy of 1.5 J. It is seen that the heat capacity of a buffer gas is not the only characteristic determining its efficiency. For helium and argon, which have similar heat capacities, the output energies are in fact different, their efficiency ratio depending on the buffer-gas pressure. Employing nitrogen seems to be highly advantageous owing to its high efficiency and low cost.

Sulfur hexafluoride SF_6 engages our attention because ofi ts high efficiency. As compared with other gases investigated, SF_6 possesses a high heat capacity ($c_p = 97$ J mol^{-1} K^{-1} for 298 K [10]), nearly three times that of nitrogen and five times that of argon. Furthermore, the sulfur hexafluoride dissociates in the discharge to produce fluorine atoms. This feature furnishes an opportunity to produce additional iodine atoms with the use of hydrogen iodide in lieu of alkyliodide molecules by the reaction

$$F + HI(DI) = I + HF(DF).$$

Therefore, there appears an opportunity to increase the iodine atom concentration by increasing the SF_6 pressure instead of the iodide pressure. This approach seems to be more efficient. Moreover, owing to the high sulfur – hexafluoride pressures, the properties of the working mixture of a pulsed COIL approach those of the mixtures of a nonchain-reaction HF laser. This gives promise that the discharge circuits employed to advantage for HF lasers [11] may be realised. Unfortunately, attempts to raise the partial pressure of the sulfur hexafluoride above 7 Torr in the experiment did not meet with success owing to the insufficient conveyance of the gas feed system.

With tetrachloride of carbon CCl_4 in lieu of sulfur hexafluoride, the chemical production of iodine atoms can be accomplished by the reaction

$$Cl + HI(DI) = I + HCl(DCl).$$

The use of this reaction is favoured because the quenching rate of singlet oxygen by HCl molecules is lower than the quenching rate by HF(DF) molecules [12]:

$$O_2(^1\Delta) + HF = O_2(^3\Sigma) + HF, \quad K = 4 \times 10^{-16} \text{ cm}^3 \text{ s}^{-1},$$

$$O_2(^1\Delta) + HCl = O_2(^3\Sigma) + HCl, \quad K = 4 \times 10^{-18} \text{ cm}^3 \text{ s}^{-1}.$$

In this case, the quenching rate of the excited iodine [2], a factor of significance for the laser operation owing to the high rate of energy transfer from the singlet oxygen molecules to the iodine atoms.

One can expect that different Freons may also serve as efficient buffer gases. This circumstance would allow choosing a diluent offering the best performance. All these suggestions evidently call for experimental verification.

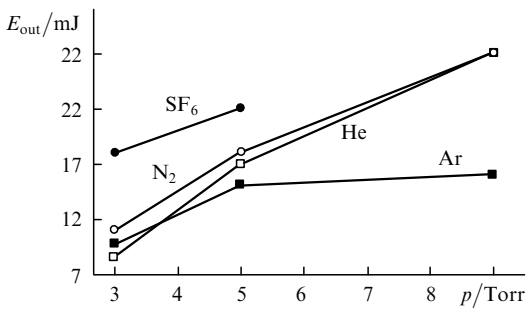

Figure 4. Dependences of the output laser energy E_{out} on the partial pressure of different buffer gases p for a pump energy 1.5 J, $p_{CH_3I} = 0.5$ Torr, $p_{O_2} = 1.0$ Torr.

4. Conclusions

The investigation of a pulsed COIL, based on a sparger SOG, initiated by a transverse self-sustained electric discharge performed in this work demonstrates that this kind of initiation holds promise for obtaining short pulses. The resultant pulse duration equal to 6.5 μs (FWHM) corresponds to an iodine atom concentration of 1.8×10^{15} cm^{-3} produced by the discharge in the laser active medium.

The lasing was obtained for a total pressure in the resonator up to 10 Torr and a partial oxygen pressure up to 2 Torr. It follows from the experimental results that a further increase of the operating pressure is possible. Sulfur hexafluoride exhibited a good performance as a buffer gas, which gives grounds to expect a further increase of the iodine atom concentration as a result of the chemical production by the $F + HI(DI) = I + HF(DF)$ reaction with the use of HI(DI) as an iodine donor. An implementation of the scheme under study would allow the development of a pulsed COIL with characteristics close to those of a photodissociation iodine laser and yet with significantly higher electric efficiency and the capacity to operate in a repetitively pulsed mode inherent in gas-flow lasers.

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