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Similarity criteria in calculations of the energy characteristics of a cw oxygen–iodine laser

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Abstract. **The calculated and experimental data on the energy efficiency of a cw oxygen–iodine laser (OIL) are analysed based on two similarity criteria, namely, on the ratio of the residence time of the gas mixture in the resonator to the characteristic time of extraction of the energy stored in singlet oxygen τ_d and on the gain-to-loss ratio Π. It is shown that the simplified two-level laser model satisfactorily predicts the output characteristics of OILs with a stable resonator at $\tau_d \leqslant 7$. Efficient energy extraction from the OIL active medium is achieved in the case of $\tau_d = 5-7$, $\Pi = 4-8$.**

Keywords: *stable resonator, singlet oxygen, oxygen–iodine laser, gain coefficient, homogeneous saturation, OIL, $O_2(^1\Delta)$.*

Oxygen–iodine lasers (OILs) belong to the class of energy transfer lasers, their remarkable feature being a high rate of electronic excitation energy exchange between energy carriers [singlet oxygen $O_2(^1\Delta)$] and emitting particles (iodine atoms),

$$O_2(^1\Delta) + I(^2P_{3/2}) \underset{k_r}{\overset{k_f}{\longleftrightarrow}} O_2(^3\Sigma) + I(^2P_{1/2}), \quad (1)$$

where $O_2(^3\Sigma)$ is the oxygen molecule in the ground state; $I(^2P_{1/2})$ and $I(^2P_{3/2})$ are the iodine atoms in the excited and ground states, respectively; $k_f = 7.8 \times 10^{-11}$ cm^3 s^{-1} [1] is the direct reaction rate constant (1); and k_r is the reverse reaction rate constant.

The oxygen–iodine mixture can amplify propagating radiation if the singlet oxygen yield in the flow $Y = [O_2(^1\Delta)]/[O_2]_0$ exceeds the threshold value determined by the expression

$$Y_{th} = \frac{1}{2K_{eq} + 1}. \quad (2)$$

Here, $K_{eq} = k_f/k_r = 0.75\exp(401.4/T)$ is the equilibrium constant of process (1), T is the temperature (in K), $[O_2(^1\Delta)]$ is the concentration of oxygen molecules in the singlet state, and $[O_2]_0$ is the total concentration of oxygen molecules. The threshold concentration of $O_2(^1\Delta)$ is $\sim$15% at room temperature and decreases with decreasing temperature.

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At present, the best developed are OILs in which singlet oxygen is produced by the chemical method of chlorination of an alkaline solution of hydrogen peroxide.

This reaction is described by the gross equation

$$Cl_2 + H_2O_2 + 2KOH \rightarrow O_2(^1\Delta) + 2KCl + 2H_2O.$$

The yield of $O_2(^1\Delta)$ in this reaction is close to 100%. In most gas–liquid generators of singlet oxygen, the degree of chlorine utilisation is high, $U = G_{O_2}/G_{Cl_2} \geqslant 90\%$, where G_{O_2} and G_{Cl_2} are the oxygen flow rate at the exit from the generator and the chlorine flow rate at the entrance to the generator.

The energy efficiency η of a cw (chemical) OIL is determined as the ratio of the output power W_{out} to the maximum possible power W_p,

$$\eta = \frac{W_{out}}{W_p} = \frac{W_{out}}{q_{ph} G_{Cl_2}}, \quad (3)$$

where q_{ph} is the laser photon energy.

A large number of works is devoted to experimental studies of the energy characteristics of cw OILs with both chemical [2–22] and electric-discharge [23–26] singlet oxygen generators. These studies were performed using wide-aperture stable resonators since they provide the best conditions for efficient energy extraction (homogeneous gain profile saturation, low diffraction losses, etc.). The values η exceeding 30% were achieved in many works [6, 11–14]. The highest parameter $\eta = 40\%$ was obtained in [11].

Papers [27–32] present the output parameters of a chemical OIL calculated numerically based on models taking into account optical, kinetic, and gas-dynamic processes. However, the simulation of mixing high-speed gas flows is complicated by simultaneous occurring of a large number of chemical and energy-exchange processes in the active medium [33, 34]. This makes it difficult to qualitatively analyse the factors affecting the output laser parameters.

In recent paper [1], the experimental output powers and gain coefficients were compared with the data calculated based on a one-dimensional model on the assumption of instantaneous mixing and on a three-dimensional model taking into account turbulent diffusion. It was shown that the one-dimensional model predicts these parameters as well as the three-dimensional model does. It was concluded that, in the case of using efficient mixing systems, the energy losses due to mixing of flows are insignificant.

Simple analytical models allowing one to determine the output characteristics of OILs with a Fabry–Perot resonator and a wide-aperture stable resonator were developed in [35–39]. All these works are based on the so-called two-level laser model, according to which the level population kinetics

is determined solely by energy-exchange process (1) and induced radiation with a frequency coinciding with the centre of the spectral line of the $I(^2P_{1/2}, F = 3) \rightarrow I(^2P_{3/2}, F' = 4)$ transition. This approach allows one to obtain a series of simple dependences of the OIL energy characteristics on several dimensionless similarity criteria [37].

In [39], the dependences of the output OIL characteristics on the resonator length along the flow were studied experimentally and numerically. It was shown that the two-level lasing model allows one to predict the OIL energy characteristics with a high accuracy. However, due to considerable simplifications used in this model, the obtained universal dependences can be used for estimation only in the cases when the energy losses in the mixing and relaxation processes are insignificant. The boundaries of the ranges of parameters of cw OILs in which the two-level model satisfactorily predicts the output energy characteristics are still not precisely determined.

In the present work, we consider a simplified generation model for a cw OIL with a stable resonator, in which we introduce dimensionless similarity criteria having a clear physical meaning. Based on the analysis of calculated and published experimental data, the regions of dimensionless similarity criteria are found for which the two-level model satisfactorily predicts the output OIL characteristics. The similarity criteria corresponding to the lasing regimes with a high energy efficiency are determined.

One of the important parameters of OILs, which in many respects determines their energy efficiency, is the yield of singlet oxygen at the entrance to the resonator Y_0. Let us transform relation (3) so that the formula for the energy (chemical) efficiency contained the experimentally measured parameter Y_0. Note that the output laser power can be represented in the form

$$W_{out} = q_{ph}(G_\Delta^0 - \Delta G_{loss} - G_\Delta^d)\eta_{extr}, \quad (4)$$

where G_Δ^0 and G_Δ^d are the flow rates of singlet oxygen at the entrance and exit of the resonator, respectively; ΔG_{loss} denotes the losses of singlet oxygen in the processes of iodine molecule dissociation and relaxation, $\eta_{extr} = t_r/(t_r + \alpha)$ is the portion of radiative energy extracted from the resonator; t_r is the total transmittance of the resonator mirrors; and α is the nonresonant loss factor. Substituting expression (4) into (3), we easily obtain the following relation for the energy efficiency of a chemical OIL:

$$\eta = U(Y_0 - \Delta Y_{loss} - Y_d)\eta_{extr}, \quad (5)$$

where ΔY_{loss} is the value of singlet oxygen losses in the resonator in the processes of I_2 dissociation and relaxation and Y_d is the yield of singlet oxygen at the exit from the resonator. It should be taken into account that expression (5) includes both the experimentally measured parameters U, Y_0, and η_{extr} and the calculated parameters ΔY_{loss} and Y_d. Note that, to find the latter parameters, it is enough to consider only the processes occurring in the lasing zone. In the general case, to find the values of ΔY_{loss} and Y_d, the calculation models must take into account the optical, kinetic, and gas-dynamic processes.

In order to obtain some general laws to describe lasing of OILs, we assume, similar to [35–39], that the active medium kinetics is completely determined by the energy-exchange process (1). From this assumption, it follows that $\Delta Y_{loss} = 0$. In addition, we assume that the intraresonator intensity is constant in the entire volume occupied by radiation and that the gain saturation is homogeneous, which is true for lasers with a wide-aperture stable resonator [35]. The collisional broadening of the gain profile becomes noticeable at the gas pressures of 10–20 Torr [40]. In lasers operating with a high energy efficiency, the pressure in the resonator is usually lower, about several Torrs [11–15].

In the nozzle block directly in front of the entrance to the resonator (Fig. 1), a secondary flow consisting of molecular iodine vapour and iodine-carrying buffer gas was admixed to the primary flow from the $O_2(^1\Delta)$ generator with a specified yield of singlet oxygen Y_0. We will assume that the resonator is placed in the flow zone where the processes of mixing and molecular iodine dissociation are completed. The temperature, the gas-mixture density, and the flow rate in this model are taken to be constant.

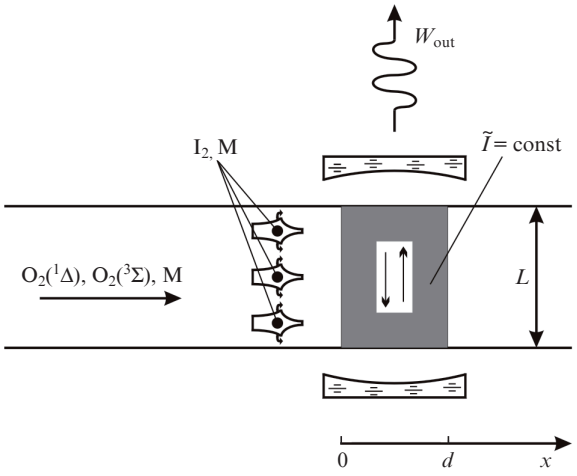

Figure 1. Principal scheme of an OIL: (M) buffer gas; (L) gain region length; (d) size of the region occupied by radiation on the mirror in the direction of the flow; (x) coordinate along the flow.

With these assumptions, the excitation kinetics and the lasing process are described by the equations for the gain coefficient g and the rates of change in the concentrations of singlet oxygen and excited atomic iodine together with the expressions for the total concentrations of molecular oxygen and atomic iodine [35–39]:

$$g = \sigma\left([I^*] - \frac{1}{2}[I]\right), \quad (6)$$

$$\frac{d[O_2(^1\Delta)]}{dt} = -k_f[I][O_2(^1\Delta)] + k_r[I^*][O_2(^3\Sigma)], \quad (7)$$

$$\frac{d[I^*]}{dt} = k_f[I][O_2(^1\Delta)] - k_r[I^*][O_2(^3\Sigma)] - \frac{g\tilde{I}}{h\nu}, \quad (8)$$

$$[O_2]_0 = [O_2(^1\Delta)] + [O_2(^3\Sigma)] = \text{const}, \quad (9)$$

$$[I]_0 = [I^*] + [I] = \text{const}, \quad (10)$$

where σ is the induced absorption/emission cross section for the Doppler-broadened transition $I(^2P_{1/2}, F = 3) \rightarrow I(^2P_{3/2},$

$F' = 4$) determined by the expression $\sigma = 1.3 \times 10^{-16}/\sqrt{T}$ cm^2; $[O_2(^3\Sigma)]$ is the concentration of oxygen molecules in the ground electronic state; $[I^*]$ and $[I]$ are the concentrations of iodine atoms in the excited $[I(^2P_{1/2})]$ and ground $[I(^2P_{3/2})]$ states, respectively; $t = x/u$ is the residence time of the gas mixture in the region occupied by radiation; x is the flow coordinate measured from the beginning of this region; u is the gas velocity; $\widetilde{I}$ is the intraresonator intensity of radiation circulating in both directions; $h\nu$ is the energy of a photon emitted at the $I(^2P_{1/2}, F = 3) \rightarrow I(^2P_{3/2}, F' = 4)$ transition with the wavelength $\lambda = 1.315$ μm.

Taking (10) into account, expression (6) can be transformed so that the gain coefficient was a function of only one variable – relative concentration of excited iodine atoms, $\eta_{I^*} = [I^*]/[I]_0$. As a result, we have

$$g = \frac{\sigma[I]_0}{2}(3\eta_{I^*} - 1). \tag{11}$$

The concentration of excited iodine atoms in the resonator can be taken to be steady-state ($d[I^*]/dt \approx 0$), since the characteristic time of energy exchange between singlet oxygen and atomic iodine is several orders of magnitude smaller than the gas time-of-flow between the mirrors. Then, taking into account (9) and (10), from (8) we obtain the following formula for calculating η_{I^*} as a function of the normalised intraresonator intensity $C = \sigma\widetilde{I}/(h\nu k_f[O_2]_0)$ and the singlet oxygen yield Y:

$$\eta_{I^*} = \frac{2K_{eq}Y + K_{eq}C}{2[(K_{eq} - 1)Y + 1] + 3K_{eq}C}. \tag{12}$$

Taking into account that the typical active medium condition is $[O_2]_0 \gg [I]_0$ and keeping in mind the above steady-state condition, we can obtain from (7) and (8) the following equation describing the rate of change in the singlet oxygen concentration in the course of interaction of induced radiation with the active medium:

$$\frac{d[O_2(^1\Delta)]}{dt} = -\frac{g\widetilde{I}}{h\nu}.$$

After simple mathematical transformations, this equation can be represented in the form containing only dimensionless parameters,

$$\frac{dY}{d\tau} = -\frac{1}{2}C(3\eta_{I^*} - 1), \tag{13}$$

where $\tau = k_f[I]_0 t$ is the ratio of the residence time of the active medium in the laser radiation field to the characteristic time of the singlet oxygen energy extraction $(k_f[I]_0)^{-1}$. Taking into account (2) and (12), this equations can be written as

$$\frac{dY}{d\tau} = \frac{C(1 - Y/Y_{th})}{2[(K_{eq} - 1)Y + 1] + 3K_{eq}C}. \tag{14}$$

The normalised intraresonator intensity C can be found from the steady-state oscillation condition, which, for a resonator with a homogeneous field, has the form ([41], p. 41)

$$\int_0^d g(x)\,dx = \frac{(t_r + \alpha)d}{2L},$$

where d is the size of the region occupied by radiation on the mirror in the flow direction (in the general case, this size does not exceed the resonator mirror dimension along the flow, see Fig. 1) and L is the gain length. In this equation, taking the gas velocity in the resonator to be constant, we perform integration over the variable τ and, taking into account (11) and (13), derive the equation

$$-\int_0^{\tau_d} \frac{\sigma[I]_0}{C}\frac{dY}{d\tau}d\tau = \frac{(t_r + \alpha)\tau_d}{2L}.$$

Finally, the steady-state oscillation condition can be written in the form

$$Y_d = Y_0 - \frac{C\tau_d}{\Pi}, \tag{15}$$

where the dimensionless similarity criterion $\Pi = 2\sigma[I]_0 L/(t_r + \alpha)$ corresponds to the gain-to-loss ratio and the similarity criterion $\tau_d = k_f[I]_0 d/u$ is the ratio of the residence time of the gas in the lasing zone $t_d = d/u$ to the characteristic time of the singlet oxygen energy extraction $(k_f[I]_0)^{-1}$ at an infinite intraresonator intensity.

The solution to differential equation (14) with the initial condition in the form of the singlet oxygen yield at the entrance to the resonator together with the steady-state oscillation condition (15) allows one to determine the singlet oxygen yield Y_d at the exit of the resonator. The dimensionless intensity C is found using an iterative procedure. The form of Eqns (14) and (15) allows us to conclude that the solution for Y_d will be completely determined by the similarity criteria τ_d and Π. Hence, the efficiency of energy extraction from the active medium – the factor in parentheses in (5) – is also characterised by these criteria. The chemical-to-laser energy conversion efficiency can be calculated using relation (5) with known U and η_{extr}.

As an example, Fig. 2 shows the calculated dependences of the singlet oxygen yield at the resonator exit Y_d on the parameter τ_d at $Y_0 = 0.6$ and $T = 300$ K for several values of the gain-to-loss ratio Π. One can see that, at $\Pi = 1$, oscillation is absent and the degree of energy extraction increases with increasing the similarity criteria τ_d and Π. For each value of Π at $\tau_d \rightarrow \infty$, the solution tends to the asymptotic value Y_d^∞, which determines the singlet oxygen yield at the exit of an infinitely long resonator. The solutions for $\tau_d \rightarrow \infty$ are shown by the horizontal segments to the right side of Fig. 2.

To obtain the equation for the horizontal asymptotes, let us transform (14) to the form convenient for integrating,

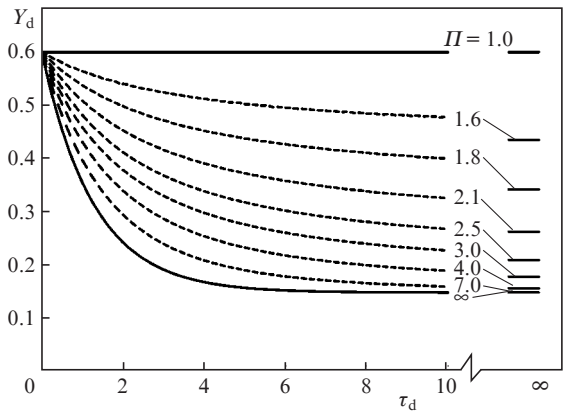

Figure 2. Dependences of the singlet oxygen yield at the exit of the cavity Y_d on the τ_d parameter at $Y_0 = 0.6$, $T = 300$ K, and different Π.

$$2Y_{th}(K_{eq}-1)\,dY + 2Y_{th}[(K_{eq}-1)\,Y_{th}+1]\frac{dY}{Y-Y_{th}}$$

$$+\frac{3K_{eq}\,Y_{th}\,C}{Y-Y_{th}}dY = -C d\tau.$$

From (15) it follows that $C|_{\tau_d\to\infty} \to 0$. In this case, the latter term in the left-hand side of the equation can be neglected. Integration from Y_0 to Y_d^∞ in the left-hand side and from 0 to τ_d in the right-hand side performed taking into account that $\Pi(Y_d^\infty - Y_0) = -C\tau_d$ allows us to obtain the following equation for horizontal asymptotes:

$$(\Pi - 1 + 3Y_{th})(Y_0 - Y_d^\infty)$$

$$-3Y_{th}(1-Y_{th})\ln\frac{Y_0-Y_{th}}{Y_d^\infty - Y_{th}} = 0. \qquad (16)$$

Equation (16), which was also obtained in [36, 38] in a somewhat different form, determines the positions of horizontal asymptotes of the curves in Fig. 2 at different Π. As follows from this transcendental equation, in the case of identical gain and loss ($\Pi = 1$), energy is not extracted from the active medium ($Y_d^\infty = Y_0$), and, at $\Pi \to \infty$, the asymptotic value is equal to the threshold singlet oxygen yield ($Y_d^\infty = Y_{th}$). Note that we can take $Y_d^\infty = Y_{th}$ even at $\tau_d \geqslant 5$ and $\Pi \geqslant 4$. This means that the losses of singlet oxygen in the resonator in these regimes are insignificant.

The lowest solid curve in Fig. 2 represents the dependence $Y_d(\tau_d)$ obtained on the strong-field assumption. Taking $C \to \infty$, we can transform Eq. (14) to the form

$$\frac{dY}{d\tau} = -\frac{Y-Y_{th}}{3K_{eq}\,Y_{th}}.$$

Integration of this equation with the initial condition $Y|_{\tau_d=0} = Y_0$ leads to the following expression for calculating Y_d in analytic form [37]:

$$Y_d = (Y_0 - Y_{th})\exp\left(-\frac{\tau_d}{3K_{eq}\,Y_{th}}\right) + Y_{th}. \qquad (17)$$

Thus, the simplified two-level model of a cw OIL makes it possible to introduce two dimensionless similarity criteria, τ_d and Π, which have clear physical meaning. These criteria include the following experimentally measured values: concentration of iodine atoms $[I]_0$, size of the region occupied by radiation on the mirror in the direction of flow d, the gas velocity u, the gain length L, the total transmittance of mirrors t_r, and the nonresonant loss coefficient α. Simple calculations allow one to estimate the energy efficiency of a laser even on the stage of its design from the given chlorine utilisation degree U, singlet oxygen yield Y_0 in the flow at the entrance to the resonator, and the portion of radiative energy extracted from the resonator η_{extr}. In addition, the simplified model yields the relations for calculating the maximum energy extraction from the active medium volume at $\tau_d \to \infty$ for any Π [Eqn (16)] and at $\Pi \to \infty$ for any τ_d [Eqn (17)].

The simplified model does not take into account the electron energy losses for molecular iodine dissociation and relaxation. The energy released in relaxation processes increases the flow temperature, which negatively affects the efficiency of energy extraction from the active medium. Figure 3 shows the dependences of the singlet oxygen yield at the exit from the resonator Y_d on the parameter τ_d at different temperatures. The initial singlet oxygen yield and the gain-to-loss ratio were taken to be Y_0 =0.6 and Π = 3. The positions of the horizontal asymptotes of the curves in Fig. 3 in this case are also determined by Eqn (16). According to the simplified model, the less efficient energy extraction from the active medium at higher temperatures is completely determined by the shift of the reaction (1) equilibrium to the left with increasing T. For example, at τ_d = 5, an increase in the temperature from 300 to 400 K leads to a decrease in the energy extraction efficiency by approximately 7%.

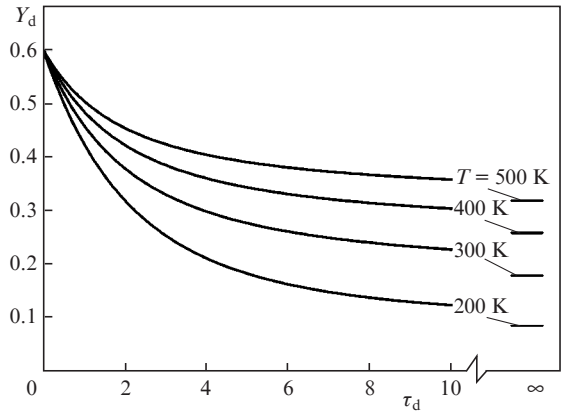

Figure 3. Dependences of the singlet oxygen yield at the exit of the cavity Y_d on the τ_d parameter at $Y_0 = 0.6$, $\Pi = 3$, and different T.

The two-level model of OIL predicts a monotonic increase in the energy extraction efficiency as the similarity criteria tend to the limiting value $Y_0 - Y_{th}$. On the other hand, an increase in the similarity criteria is accompanied by an increase in the energy losses in relaxation processes, which is not taken into account in the chosen simplified model. In reality, with increasing τ_d and Π, the efficiency of energy extraction from the active medium first reaches a maximum and then decreases. A high chemical-to-laser energy conversion efficiency is achieved in the region of optimal τ_d and Π criteria close to the values corresponding to this maximum.

The allowance for relaxation processes inevitably complicates the model, and the energy efficiency in this case cannot be represented as a function of τ_d and Π. The energy efficiency experimentally achieved in [11] was close to the limiting efficiency (η = 40%) calculated based on the two-level model. The results of this work testify to the existence of regimes in which the energy losses in relaxation processes are insignificant. Therefore, the simplified oscillation model can be used for calculating the output characteristics of cw OILs in the cases when the energy losses in relaxation processes are reduced to minimum.

The intervals of the similarity criteria corresponding to the maximum energy extraction from the active medium can be found based on a comparative analysis of the calculated and experimental data. The data obtained using the simplified oscillation model show that, at $\tau_d \geqslant 5$ and $\Pi \geqslant 4$, the energy extraction efficiency is close to the limiting value $Y_0 - Y_{th}$. The chosen model allows one to determine only the lower boundaries of the regions of similarity criteria, while the upper boundaries can be determined by analysing the output OIL characteristics available in experimental works.

Table 1. Parameters characterising the operation of OILs in experiments demonstrating a high chemical efficiency.

U	Y_0	$[I]_0$/cm^{-3}	T/K	P_{res}/Torr	u/cm s^{-1}	L/cm	d/cm	t_r	α	η_{extr}	η_{exp} (%)	Paper
0.94	0,68	7.5×10^{14}	280[1)]	2.3[1)]	4.2×10^4	5.0	3.6[1)]	0.007[1)]	0.0006[1)]	0.92	39.6	[11]
0.90	0.70	4.1×10^{14}	268	3.0[2)]	3.1×10^4	7.5	5.0[2)]	0.016	0.0018	0.90	32.9	[12]
0.95	0.60	7.0×10^{14}	220	1.8	6.4×10^4	37.0	6.0	0.1	0.01	0.91	31.5	[13]
0.99[3)]	0.65[3)]	0.5×10^{14}	350[3)]	0.25	0.7×10^4	100.0	7.0	0.01	0.001[3)]	0.91	30.0	[14]
0.92	0.60	6.2×10^{14}	360	1.3	3.2×10^4	10.0	4.5	0.013	0.0013	0.91	29.0	[15]
0.93	0.60	14.4×10^{14}	220[4)]	7.1	4.5×10^4	5.0	4.5	0.037	0.0037[4)]	0.91	25.6	[16]

[1)] B.D. Barmashenko (private communication), [2)] M. Endo (private communication), [3)] N.N. Yuryshev (private communication), [4)] M.V. Zagidullin (private communication).

Table 1 lists the laser parameters taken from experimental papers reporting on the achievement of a high chemical efficiency [11–16]. Additional information concerning these experiments can be also found in [17–22]. From all the references, we chose only the works containing data necessary for our analysis. Some missing parameters were directly provided by the authors of these papers. In [16], the singlet oxygen yield at the entrance to the resonator Y_0 was not measured. Here, we take it to be 0.6 – a typical value for OILs with a chemical singlet oxygen generator.

An important characteristics appearing in the chosen model and exerting a pronounced effect on the results of experiments and calculations is the concentration of iodine atoms. Under experimental conditions, this concentration is not equal to the doubled concentration of iodine molecules, because they dissociate not completely. The dissociation degree $F_{I_2} = G_{I_0}(2G_{I_2})^{-1}$ is usually 50%–80% (G_{I_0} and G_{I_2} are the flow rate of atomic iodine at the entrance to the resonator and the total flow rate of molecular iodine, respectively). The concentration of iodine atoms in the flow can be most precisely determined if the gain coefficient is know. In this case, this concentration can be determined using the relation [18]

$$[I]_0 = \frac{2g}{\sigma}\frac{K_{eq}-1}{2K_{eq}+1}\frac{Y_0+1/(K_{eq}-1)}{Y_0-Y_{th}}.$$

By this formula, the atomic iodine concentration was determined under the conditions of works [11–13], in which the gain coefficients were directly measured. The atomic concentration $[I]_0$ in [14] was the same as in work [22] by the same authors. For papers [15, 16], the atomic iodine concentration was calculated taking into account the data of these works on the relation of molecular iodine and oxygen flows on the assumption that the molecular iodine dissociation degree is 0.8 for a rake-type iodine injector [15] and 0.6 for an ejector mixing system [16].

Based on the parameters given in Table 1, we determined the similarity criteria τ_d and Π and calculated the energy efficiencies η_{cal} by solving systems of equations (14) and (15) with the use of (5). The values of τ_d and Π given in Table 2 are determined with an accuracy of ~20% due to the errors in the measurement of atomic iodine concentrations and gas temperature. As is seen from Table 2, the calculated energy efficiencies are higher than the experimental values η_{exp} on average by 6%. This occurs because the energy losses in the I_2 dissociation and relaxation processes were not taken into account.

Comparison of the calculated and experimental data shows that the losses of singlet oxygen ΔY_{loss} in the experiments chosen by us for the analysis varies from 3% to 16% (see Table 2). Since the dissociation of one iodine molecule involves three singlet oxygen molecules [34], the amount of $O_2(^1\Delta)$ molecules spent on iodine dissociation can be determined from the relation $\Delta Y_{diss} = 3\eta_{I_2}F_{I_2}$, where $\eta_{I_2} = G_{I_2}/G_{O_2}$ is the relative initial concentration of I_2 in the gas flow. From the values of ΔY_{diss} given in Table 2, it follows that the portion of singlet oxygen molecules spent on the dissociation amounts to 6%. The other part of losses, $\Delta Y_{relax} = \Delta Y_{loss} - \Delta Y_{diss}$, can be attributed to the energy losses due to the relaxation of excited $I(^2P_{1/2})$ atoms on H_2O, I_2, and $O_2(^1\Delta)$ molecules [33, 34]. For example, in [11] the dissociation losses are 4% and the relaxation losses are 1%. In the considered experimental works, except for [16], the losses of singlet oxygen in relaxation processes do not exceed several percents. It should also be noted that the main contribution to the total losses of singlet oxygen is made by dissociation losses in the case of the ultrasonic regime and by relaxation losses in the subsonic regime. This occurs because the optimal relative concentration of I_2 in the ultrasonic flow is higher than in the subsonic flow.

The two-level model predicts that, at $\tau_d < 5$ and $\Pi < 4$, the portion of singlet oxygen carried out of the resonator with the gas flow, $\Delta Y_{out} = Y_d - Y_{th}$, is rather large. In particular, in [12], the gain-to-loss ratio is small, $\Pi = 2.7$ (see Table 2), and, as a result, the outflow losses are high, $\Delta Y_{out} = 12\%$. These losses are also high ($\Delta Y_{out} = 9\%$) in work [14], where the similarity criterion $\tau_d = 3.9$ is nonoptimal. However, the nature of these losses in [12] and [14] is different, namely, in the first case, the active medium is characterised by a low gain coefficient, while in the second case, the small resonator length along the flow does not allow efficient energy extraction from the active medium. In [11, 13, 15], where $\tau_d > 5$ and $\Pi > 4$, the portion of singlet oxygen carried out of the resonator does not exceed several percents. In [16], the energy efficiency is relatively low (25.6%), which, at $\tau_d = 11.2$ and $\Pi = 3.1$, is caused mainly by the relaxation losses $\Delta Y_{relax} = 0.11$.

Table 2. Calculated parameters characterising the operation of highly efficient OILs.

Y_{th}	Y_d^∞	Y_d	τ_d	Π	η_{cal} (%)	$\Delta\eta$ (%)	η_{I_2}	F_{I_2}	ΔY_{diss}	ΔY_{relax}	ΔY_{loss}	Paper
0.14	0.14	0.17	5.0	7.7	43.6	4.0	0.027	0.48	0.04	0.01	0.05	[11]
0.13	0.15	0.25	5.1	2.7	35.3	2.4	0.014	0.64	0.03	0.00	0.03	[12]
0.10	0.10	0.14	5.1	4.1	37.6	6.1	0.025	0.74	0.06	0.01	0.07	[13]
0.17	0.18	0.26	3.9	6.3	36.4	6.4	0.004	0.80	0.01	0.06	0.07	[14]
0.18	0.18	0.22	6.8	5.9	32.2	3.2	0.017	0.80	0.04	0	0.04	[15]
0.10	0.10	0.14	11.2	3.1	39.2	13.6	0.029	0.60	0.05	0.11	0.16	[16]

Analysis of the data given in Tables 1 and 2 shows that the dimensionless similarity criteria in the studies achieved a high energy efficiency ($\eta_{exp} \geqslant 30\%$) are $_d = 5-7$ and $\Pi = 3-8$. At the same time, correction of the calculated initial singlet oxygen yield Y_0 for dissociation losses decreases the difference between the calculated and experimental energy efficiencies to several percents. In this case, $\Delta\eta = \eta_{cal} - \eta_{exp}$ corresponds solely to relaxation losses. Therefore, the simplified two-level model allows one to predict the output characteristics of cw OILs with a satisfactory accuracy at $\tau_d \leqslant 7$. The latter is true when the losses for molecular iodine dissociation are taken into account.

Generalising the calculated experimental results, we can conclude that the efficient energy extraction from the active medium of a chemical OIL is achieved at $\tau_d = 5-7$ and $\Pi = 4-8$. Here, the lower boundaries correspond to the calculated data. The upper boundaries are determined based on experimental data of papers in which high energy efficiency was achieved. The right-hand boundary chosen for the first criterion, $\tau_d = 7$, is close to the value obtained in [15], where the chemical efficiency was $\sim$30%.

Thus, the two-level model proposed to describe a cw OIL operation [35–39] allows one to introduce dimensionless similarity criteria, which have a clear physical meaning and include experimentally measured parameters. The criterion $\tau_d = k_f[I]_0 d/u$ is determined by the ratio of the residence time of the gas flow in the lasing zone to the characteristic time of singlet oxygen energy extraction. The criterion $\Pi = 2\sigma[I]_0 L/(t_r + \alpha)$ corresponds to the gain-to-loss ratio. The simplified model allows one to estimate the energy efficiency of a laser even at the stage of its creation at $\tau_d \leqslant 7$ and provides relations for calculating maximum possible energy extraction from the active medium volume at $\tau_d \to \infty$ for any Π (16) and at $\Pi \to \infty$ for any τ_d (17).

The analysis of the calculated and experimental results shows that the efficient energy extraction from the active medium of a chemical OIL is achieved at $\tau_d = 5-7$ and $\Pi = 4-8$. At lower τ_d and Π, a considerable part of energy is carried out of the resonator with the gas flow, while the relaxation losses in the active medium and the nonresonant losses at the mirrors become considerable at larger values of these parameters. At long residence times, the heat release losses also increase. An increase in temperature leads to a shift of reaction (1) equilibrium to the left and to a decrease in the efficiency of energy extraction from the active medium. The two-level lasing model can be used for estimating the output characteristics of OILs if the energy losses in the mixing and relaxation processes are insignificant.

The simplified model can also be used in calculations of the output characteristics of cw OILs with an electric-discharge singlet oxygen generator [23–26], because the similarity criteria τ_d and Π include parameters common for lasers of both types. In this case, the energy efficiency is determined by expression (5), in which the factor U is taken to be equal to unity.

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References

1. Brami-Rosilio I., Barmashenko B.D., Rosenwaks S. *Appl. Phys. B*, **108**, 615 (2012).
2. McDermott W.E., Stephens J.C., Vetrovec J., Dickerson R.A. *AIAA 97-2385, 28th Plasmadynamics and Lasers Conf.* (Atlanta, GA, 1997).
3. Azyazov V.N., Zagidullin M.V., Nikolaev V.D., Svistun M.I., Khvatov N.A. *Kvantovaya Elektron.*, **22** (5), 443 (1995) [*Quantum Electron.*, **25** (5), 418 (1995)].
4. Rybalkin V., Katz A., Barmashenko B.D., Rosenwaks S. *Appl. Phys. Lett.*, **82**, 3838 (2003).
5. Zagidullin M.V., Nikolaev V.D., Svistun M.I., Khvatov N.A. *Kvantovaya Elektron.*, **35** (10), 907 (2005) [*Quantum Electron.*, **35** (10), 907 (2005)].
6. Adamenkov A.A., Bakshin V.V., Bakshin A.V., et al. *Kvantovaya Elektron.*, **37** (7), 601 (2007) [*Quantum Electron.*, **37** (7), 601 (2007)].
7. Truesdell K.A. *Proc. SPIE Int. Soc. Opt. Eng.*, **6346**, 63461L (2007).
8. Xu M., Sang F., et al. *Jpn. J. Appl. Phys.*, **47**, 8446 (2008).
9. Rajesh R., Singhal G., Mainuddin, Tyagi R.K., Dawar A.L. *Opt. & Laser Technol.*, **42**, 580 (2010).
10. Mikheyev P.A., Zagidullin M.V., Azyazov V.N. *Appl. Phys. B*, **101**, 7 (2010).
11. Rybalkin V., Katz A., Barmashenko B.D., Rosenwaks S. *Appl. Phys. Lett.*, **85**, 5851 (2004).
12. Endo M., Osaka T., Takeda S. *Appl. Phys. Lett.*, **84**, 2983 (2004).
13. Boreisho A.S., Barkan A.B., Vasil'ev D.N., Evdokimov I.M., Savin A.V. *Kvantovaya Elektron.*, **35** (6), 495 (2005) [*Quantum Electron.*, **35** (6), 495 (2005)].
14. Vagin N.P., Karapetyan D.G., Konoshenko A.F., Kryukov P.G., Pazyuk V.S., Tomashov V.N., Yuryshev N.N. *Kr. Soobshch. Fiz. FIAN*, (4), 6 (1989).
15. Antonov I.O., Azyazov V.N., Mezhenin A.V., Popkov G.N., Ufimtsev N.I. *Appl. Phys. Lett.*, **89**, 051115 (2006).
16. Nikolaev V.D., Svistun M.I., Zagidullin M.V., Hager G.D. *Appl. Phys. Lett.*, **86**, 231102 (2005).
17. Rybalkin V., Katz A., Barmashenko B.D., Rosenwaks S. *J. Appl. Phys.*, **98**, 023106 (2005).
18. Waichman K., Barmashenko B.D., Rosenwaks S. *J. Chem. Phys.*, **133**, 084301 (2010).
19. Endo M., Masuda T., Uchiyama T. *IEEE J. Quantum Electron.*, **42**, 71 (2006).
20. Endo M., Masuda T., Uchiyama T. *AIAA J.*, **45**, 90 (2007).
21. Boreisho A.S., Mal'kov V.M., Savin A.V., Vasil'ev D.N., Evdokimov I.M., et al. *Kvantovaya Elektron.*, **33** (4), 307 (2003) [*Quantum Electron.*, **33** (4), 307 (2003)].
22. Konoshenko A.F., Kryukov P.G., Nurligareev D.Kh., Pazyuk V.S., Tomashov V.N., Yuryshev N.N., Vagin N.P., in *Proc. of the First International Workshop on Iodine Laser and Applications* (Bechynê: Inst. of Phys. CAS, 1986) p. 253.
23. Woodard B.S., Benavides G.F., Zimmerman J.W., Carroll D.L., Palla A.D., Day M.T., et al. *Opt. Lett.*, **35**, 1611 (2010).
24. Woodard B.S., Zimmerman J.W., Benavides G.F., Carroll D.L., Verdeyen J.T., Palla A.D., Field T.H., Solomon W.C., Lee S., Rawlins W.T., Davis S.J. *J. Phys. D: Appl. Phys.*, **43**, 025208 (2010).
25. Zimmerman J.W., Benavides G.F., Palla A.D., Woodard B.S., et al. *Appl. Phys. Lett.*, **94**, 021109 (2009).
26. Benavides G.F., Zimmerman J.W., Woodard B.S., Carroll D.L., Verdeyen J.T., et al. *Appl. Phys. Lett.*, **92**, 041116 (2008).
27. Palla A.D., Carroll D.L., Solomon W.C. *AIAA 2011–4006, 42nd Plasmadynamics and Lasers Conf.* (Honolulu, Hawii, 2011).
28. Carroll D.L. *AIAA J.*, **34**, 338 (1996).
29. Yang T.T., Copeland D.A., Bauer A.H., Qaun V., McDermott W.E., Cover R.A., Smith D.M. *AIAA 97-2384, 28th Plasmadynamics and Lasers Conf.* (Atlanta, GA, 1997).
30. Paschkewitz J., Shang J., Miller J., Madden T. *AIAA 2000–2574, 31st Plasmadynamics and Lasers Conf.* (Denver, CO, 2000).
31. Madden T.J. *Proc. SPIE Int. Soc. Opt. Eng.*, **6346**, 634620 (2006).
32. Waichman K., Barmashenko B.D., Rosenwaks S. *J. Appl. Phys.*, **106**, 063108 (2009).

33. Azyazov V.N. *Kvantovaya Elektron*., **39** (11), 989 (2009) [*Quantum Electron*., **39** (11), 989 (2009)].
34. Azyazov V.N., Pichugin S.Yu., Heaven M.C. *J. Chem. Phys*., **130**, 104306 (2009).
35. Hager G.D., Helms C.A., Truesdell K.A., Plummer D., Erkkila J., Growell P. *IEEE J. Quantum Electron*., **32**, 1525 (1996).
36. Barmashenko B.D., Rosenwaks S. *Appl. Opt*., **35**, 7091 (1996).
37. Barmashenko B.D., Furman D., Rosenwaks S. *Appl. Opt*., **37**, 5697 (1998).
38. Zagidullin M.V., Igoshin V.I., Katulin V.A., Kupriyanov N.L. *Preprint FIAN, No. 271* (Moscow, 1982).
39. Zagidullin M.V., Nikolaev V.D. *Kvantovaya Elektron*., **24** (5), 423 (1997) [*Quantum Electron*., **27** (5), 411 (1997)].
40. Yuryshev N.N. *Kvantovaya Elektron*., **23** (7), 583 (1996) [*Quantum Electron*., **26** (7), 567 (1996)].
41. Losev S.A. *Gazodinamichesckie lasery* (Gas-Dynamic Lasers) (Moscow: Nauka (1977).