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Power Dependence of Chemical Oxygen–Iodine Lasers on Iodine Dissociation

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The loss of $O_2(^1\Delta)$ during iodine dissociation in the chemical oxygen–iodine laser (COIL) is one of the main factors affecting the output power. Analytical expression is obtained for the number of $O_2(^1\Delta)$ molecules, N, lost in the region of I_2 dissociation per one molecule of I_2. This expression yields $N = 4$–6, in agreement with numerical calculations and experimental measurements. It is shown that some effective number $N_1 < N$ should be used rather than N to calculate the power. Analytical expression for the power is obtained, taking into account the $O_2(^1\Delta)$ losses in the dissociation region. It is shown that N increases and the power decreases when the dissociation fraction F increases. Therefore, maximum power is achieved at low values of the iodine flow rate when iodine is not completely dissociated before the resonator and when the small signal gain in the resonator region is less than its maximum achievable value. Numerical modeling of the RADICL (supersonic COIL) is carried out. The values of the $O_2(^1\Delta)$ yield and of the mixing rate are estimated to reach an agreement between the calculated and the measured dependencies of the power on the iodine flow rate.

Nomenclature

c = empirical factor
D = diffusion coefficient
F = iodine dissociation fraction
f = factor given by Eq. (17)
g_{th} = threshold gain
H = height of main flow
h_j = height of iodine jet
K_e = equilibrium constant of reaction 1
k_i = rate constant of reaction i
l_{dis} = length of dissociation region
N = number of $O_2(^1\Delta)$ molecules lost in the region of I_2 dissociation per one I_2 molecule
N_1 = effective number N used to calculate the power
N_0 = value of N_1 in the ideal resonator with $g_{th} = 0$
nZ = flow rate of species Z ($Z = I_2, O_2, He, H_2O$)
p = pressure
T = temperature
U_m = velocity of main flow
W_1 = output power
x_{ind} = induction interval
x_{res} = distance between injection point and closest end of the resonator
Y = $O_2(^1\Delta)$ yield
Y_{dis} = $O_2(^1\Delta)$ loss during dissociation
Y_{th} = threshold yield in a Fabry–Perot resonator
$[Z]$ = number density of species Z ($Z = I_2, O_2, He, H_2O$)
β = nI_2/nO_2, titration ratio
ε = $g_{th}HU_m/(2\sigma nI_2)$, ratio between g_{th} and the maximum possible small signal gain
η_{dis} = dissociation efficiency
η_{mix} = $h_j(x_{res})/H$, mixing efficiency
σ = cross section of stimulated emission
χ = nH_2O/nO_2, water ratio
ψ = nHe/nO_2, helium ratio

Subscripts

c = critical flow rate (corresponding to maximum gain)
j = jet
m = main flow
max = maximum power
th = threshold

Superscript

$*$ = excited I and I_2

I. Introduction

IN the chemical oxygen–iodine laser (COIL), iodine molecules admixed to the flow of singlet oxygen, $O_2(^1\Delta)$, are dissociated to atoms because of collisions with the $O_2(^1\Delta)$ molecules. An important parameter of dissociation, which affects the output power, is the number of $O_2(^1\Delta)$ molecules, N, lost in the region of I_2 dissociation per molecule of I_2. The parameter N determines what fraction of the energy contained in $O_2(^1\Delta)$ and available for lasing is lost in the I_2 dissociation region. From energy considerations, N must be at least two[1]; however, numerical computations[1–3] and experimental measurements[4] show that N is in the range of 4–6. It is very important to know the exact value of N to estimate the power when one applies the heuristic approach of Hon et al.[1] The only analytical expression for N is derived by Crowell and Plummer[2] [see Eq. (72) in Ref. 2]. However, they had to incorporate into the expression some unexplained empirical factor of 2.5 to reach an agreement with numerical computations. We derive an analytical expression for N without any arbitrary assumption. Our expression is different from theirs. N is found for both premixed flow and flow with finite mixing rate between the oxygen and iodine. For the latter case, the model presented previously[5] is used. It appears that N comprises the losses of $O_2(^1\Delta)$ for I_2 dissociation, quenching of $O_2(^1\Delta)$ and I^* ($I^* \equiv I(^2P_{1/2})$) and excitation of I atoms produced via the iodine dissociation. The energy of $O_2(^1\Delta)$ lost for excitation of I is contained in I^* and some part of it can be converted into the laser energy in the resonator. We show that, to calculate the power, one should use some effective number $N_1 < N$ rather than N. The expression for N_1 also is derived. The dependence of N on different parameters [iodine flow rate, $O_2(^1\Delta)$ yield, and H_2O molar fraction] is studied.

The expression for N is used to determine the losses of the energy carried by $O_2(^1\Delta)$ before the resonator and the power as a function of the iodine flow rate, nI_2. Using our model of the COIL,[5] which accounts for the mixing effects, an analytical expression is found for the power extracted from Fabry–Perot resonators. This expression is very similar to that obtained in the heuristic model of the COIL.[1] The power appears to be a nonmonotonic function of nI_2. There is

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an optimal value of $nI_2 \equiv (nI_2)_{\max}$ for which the power achieves its maximum. The maximum iodine flow rate $(nI_2)_{\max}$ appears to be much smaller than the critical flow rate $(nI_2)_c$. The latter was introduced previously[5] and corresponds to the maximum of the gain and the minimum of the iodine dissociation length. The iodine dissociation fraction F in the resonator corresponding to $(nI_2)_{\max}$ is smaller than 100%. The value of F depends on the relation between the small signal gain g_0 and the threshold gain g_{th}. It is shown below that if g_0 is 2–3 times greater than g_{th}, F is about 80%. However, if g_0 is 8–10 times greater than g_{th}, F is about 60%. This is in contrast to the common statement that the highest power is achieved only if the iodine is almost completely dissociated before the resonator. The greater the ratio g_0/g_{th}, the smaller F is.

To check these conclusions, dependencies of the power and dissociation fraction on the iodine flow rate in the case of RADICL (the supersonic COIL described by Helms et al.[6]) were calculated numerically and compared with experimental measurements.

II. Parameter *N* for Flow in a Channel with Constant Cross Section

An analytic expression for N can be obtained for flow in a channel with constant cross section. The model described previously[5] is used. The initial conditions correspond to the injection of a plannar I_2/He jet of very small initial height into the main $O_2/O_2(^1\Delta)$/He flow so that the velocities of the injected and main flows are parallel. As shown previously,[5] these initial conditions also describe well the case of transverse iodine injection. The model uses the set of chemical reactions presented in Table 1 and is based on the mechanism suggested by Heidner et al.[7] The temperature dependence of the rate constant k_{1f} of the pumping reaction 1 is different from that used elsewhere.[5,8–13] This dependence has recently been suggested by Van Marter et al.[14] The rate constant k_3 determining the rate of iodine dissociation (reaction 3) is assumed here to be independent of the temperature. Note that many authors used strong temperature dependence of k_3 (Refs. 5, 8–13). However, recent measurements of k_3 have shown that it is really almost independent of the temperature.[15,16] We use $k_3 = 3 \times 10^{-11}$ cm^3/s, which is a little smaller than $k_3 = 3.5 \times 10^{-11}$ cm^3/s used elsewhere.[2,3,7,8] Our value of k_3 is in better agreement with the measurements performed by Cline and Leone[15] and Burde et al.[16] than the old value. Indeed, $k_3 \approx 3 \times 10^{-11}$ cm^3/s, almost independent of the temperature, was measured by Cline and Leone.[15] Burde et al.[16] show that $k_3 = 3.7 \times 10^{-11}$ cm^3/s at room temperature and should decrease to about 2.3×10^{-11} cm^3/s at $T = 200$ K; hence, the average value in the COIL is about 3×10^{-11} cm^3/s. The values of k_5–k_7 in Table 1 are those reported by Heaven[17]; they are different from those previously suggested by Heidner et al.[7]

Under some approximations, it is possible to obtain an analytical solution for N. The approximations are the same as those employed previously[5]: equal velocities of the main and injected flows, constant temperature, and negligibly small rates of reactions 10–12. Only the case of low iodine flow rate, $nI_2 \ll (nI_2)_c$, is discussed. In this case, $[I_2]$ inside the iodine jet is much smaller than $[O_2]$ in the main flow in the region where F is significant (more than a few percent).

The changes of the total number density of excited particles inside the jets, $[O_2(^1\Delta)]_j + [I^*]_j$, and $[I_2]_j$ are given by

$$\begin{aligned} U_m \frac{\mathrm{d}\left(\left[O_2(^1\Delta)\right]_j + [I^*]_j\right)h_j}{\mathrm{d}x} &= U_m\left[O_2(^1\Delta)\right]_m \frac{\mathrm{d}h_j}{\mathrm{d}x} \\ &- h_j(1+\eta_{\text{dis}})\left(k_2\left[O_2(^1\Delta)\right]_j + k_3[I^*]_j\right)[I_2]_j \\ &- h_j\left(k_8[H_2O]_j + k_9\left[O_2(^1\Delta)\right]_j\right)[I^*]_j \end{aligned} \tag{1}$$

$$U_m \frac{\mathrm{d}[I_2]_j h_j}{\mathrm{d}x} = -h_j \eta_{\text{dis}}\left(k_2\left[O_2(^1\Delta)\right]_j + k_3[I^*]_j\right)[I_2]_j \tag{2}$$

where

$$\eta_{\text{dis}} = \frac{k_4 Y_m}{k_4 Y_m + k_5(1 - Y_m) + k_6\chi + k_7\psi} \tag{3}$$

is the dissociation efficiency[5] characterizing which fraction of I_2^* dissociates, $Y_m \equiv [O_2(^1\Delta)]_m/[O_2]_m$ is the yield of $O_2(^1\Delta)$, $\chi \equiv nH_2O/nO_2$, and $\psi \equiv n\text{He}/nO_2$. Unlike in earlier work,[5] we do not assume here that $\eta_{\text{dis}} = 1$ and do not neglect the quenching reactions 8 and 9. $[I^*]_j$ can be expressed through $Y_j \approx Y_m$:

$$[I^*]_j = \frac{nI_2}{h_j U_m} F \frac{2K_e Y_m}{(K_e - 1)Y_m + 1} \tag{4}$$

where

$$F \equiv 1 - \frac{[I_2]_j h_j U_m}{nI_2} \tag{5}$$

is the iodine dissociation fraction.

N can be found for both premixed flow and flow with finite mixing rate between the oxygen and the iodine. In the case of premixed flow in a channel of height H (Fig. 1a), the jet height h_j is equal to H. Therefore, the first term on the right-hand side of Eq. (1) is equal to zero. Dividing Eq. (1) by Eq. (2), one gets the following differential equation for Y as a function of F (the subscripts j and m are omitted):

$$\begin{aligned} &\frac{\mathrm{d}\{Y + 2\beta F[(K_e Y)/[(K_e - 1)Y + 1]\}}{\mathrm{d}F} \\ &\quad = -\left(1 + \frac{1}{\eta_{\text{dis}}}\right)\beta - \frac{k_6\chi + k_7 Y}{\eta_{\text{dis}} k_3(1 - F)} \end{aligned} \tag{6}$$

where $\beta \equiv nI_2/nO_2$ is the titration ratio. Equation (6) has to be solved with the initial condition $Y = Y_i$ when $F = 0$. Usually $\beta \ll 1$ and Y changes very little during dissociation. In this case, Eq. (6) can be integrated easily:

$$\begin{aligned} &Y - Y_i + 2\beta F \frac{K_e Y_i}{(K_e - 1)Y_i + 1} \\ &\quad = -\left(1 + \frac{1}{\eta_{\text{dis}}}\right)\beta F - \frac{k_6\chi + k_7 Y}{\eta_{\text{dis}} k_3} \ell n(1 - F) \end{aligned} \tag{7}$$

Table 1 Rate constants of reactions in the COIL[a]

No.	Reaction[a]	Rate constant, cm^3/mol-s	Ref.
1	$O_2(^1\Delta) + I \leftrightarrow O_2(^3\Sigma) + I^*$	$k_{1f} = 7.8 \times 10^{-11}(T/295)^{1/2}$ $k_{1b} = k_{1f}/K_e$ $K_e = 0.75\exp(402/T)$	14
2	$O_2(^1\Delta) + I_2 \rightarrow O_2(^3\Sigma) + I_2^*$	$k_2 = 7 \times 10^{-15}$	8
3	$I^* + I_2 \rightarrow I + I_2^*$	$k_3 = 3 \times 10^{-11}$	15,16
4	$I_2^* + O_2(^1\Delta) \rightarrow 2I + O_2(^3\Sigma)$	$k_4 = 3 \times 10^{-10}$	8
5	$I_2^* + O_2(^3\Sigma) \rightarrow I_2 + O_2(^3\Sigma)$	$k_5 = 5 \times 10^{-12}$	17
6	$I_2^* + H_2O \rightarrow I_2 + H_2O$	$k_6 = 1.5 \times 10^{-12}$	17
7	$I_2^* + \text{He} \rightarrow I_2 + \text{He}$	$k_7 = 1 \times 10^{-11}$	17
8	$I^* + H_2O \rightarrow I + H_2O$	$k_8 = 2 \times 10^{-12}$	8
9	$I^* + O_2(^1\Delta) \rightarrow I + O_2(^1\Sigma)$	$k_9 = 4 \times 10^{-24} T^{3.8} \exp(700/T)$	8
10	$2O_2(^1\Delta) \rightarrow O_2(^3\Sigma) + O_2(^1\Sigma)$	$k_{10} = 9.5 \times 10^{-28} T^{3.8} \exp(700/T)$	8
11	$O_2(^1\Sigma) + H_2O \rightarrow O_2(^1\Delta) + H_2O$	$k_{11} = 6.7 \times 10^{-12}$	8
12	$O_2(^1\Sigma) + I_2 \rightarrow O_2(^3\Sigma) + 2I$	$k_{12} = 4 \times 10^{-12}$	8

[a] $I^* \equiv I(^2P_{1/2})$, $I \cong I(^2P_{3/2})$ and I_2^* denotes a vibrationally excited iodine molecule.

Taking into account the definition of N,

$$N = (Y_i - Y)/\beta F \tag{8}$$

one gets finally the expression for N:

$$N = \left(1 + \frac{1}{\eta_{\text{dis}}}\right) - \frac{k_8\chi + k_9 Y_i}{\eta_{\text{dis}} k_3 \beta}\frac{\ln(1-F)}{F} + 2\frac{K_e Y_i}{(K_e - 1)Y_i + 1} \tag{9}$$

The first term on the right-hand side of Eq. (9) corresponds to the losses of $O_2(^1\Delta)$ for I_2 dissociation, the second term to the quenching of electronic excitation in reactions 6 and 7, and the third term to the excitation of iodine atoms produced via the iodine dissociation.

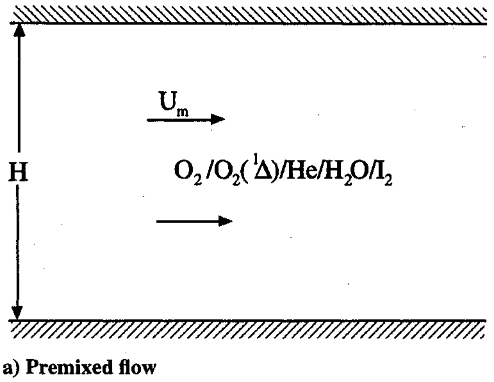
Compare the above expression for N with that derived by Crowell and Plummer.[2] See Eq. (72) in Ref. 2, which (for $\beta \ll 1$) can be rewritten in terms of η_{dis} as

$$N_{(\text{Ref. 2})} = \frac{1}{\eta_{\text{dis}}} - \frac{k_8\chi}{\eta_{\text{dis}} k_3 \beta}\frac{\ln(1-F)}{F} + 2.5 \tag{10}$$

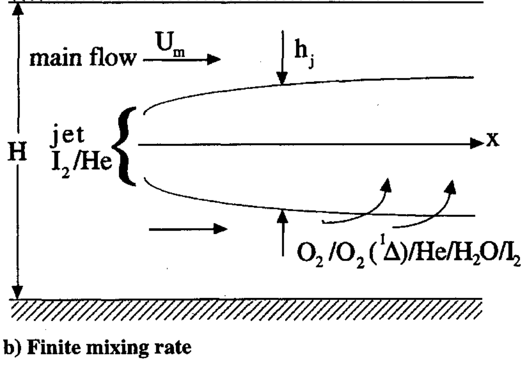
The number 2.5 on the right-hand side of Eq. (10) is an empirical correction introduced by Crowell and Plummer[2] to reach an agreement with numerical computations. It is seen that Eq. (9) for N differs from Eq. (10) for $N_{(\text{Ref. 2})}$. Reaction 9 of Table 1 is not taken into account by Crowell and Plummer[2]; thus, the term containing k_9 is absent in Eq. (10). Besides, to make the two expressions identical (neglecting reaction 9 of Table 1), the empirical factor 2.5 in Eq. (10) should be replaced with $1 + 2K_eY_i/[1 + (K_e - 1)Y_i]$. For the supersonic COIL, the typical values of the parameters are $Y_i = 0.4$, $T = 180$ K, and $K_e = 7$. Hence, $1 + 2K_eY_i/[1 + (K_e - 1)Y_i] \approx 2.65$, which is very close to the empirical factor 2.5. Therefore, our expression (9) for N explains why the empirical factor of 2.5 should be incorporated into $N_{(\text{Ref. 2})}$. Equation (9) is more general than Eq. (10) because it gives the value of N in a wide range of the parameters Y_i and T.

Consider now the case of finite mixing rate when $h_j < H$, i.e., the jet boundaries do not reach the channel walls (Fig. 1b); h_j is given by[5]

$$h_j \approx 2c\sqrt{Dx/U_m} \tag{11}$$



a) Premixed flow



b) Finite mixing rate

Fig. 1 Schematic drawing of the flow.

In Ref. 5, c was shown to be equal to 0.8 for the planar jet. In what follows, we use greater values of c to take into account the higher mixing rate in the case of transverse injection.

The definition of N differs from that given by Eq. (8) because, initially, there is no $O_2(^1\Delta)$ in the jet and the $O_2(^1\Delta)$ losses at a distance x from the injection point are equal to the difference between the fluxes of $O_2(^1\Delta)$ flowing inside the jet and coming into the jet from the main flow:

$$N = \frac{U_m h_j\left([O_2(^1\Delta)]_m - [O_2(^1\Delta)]_j\right)}{nI_2 F} \tag{12}$$

To find N, let us subtract Eq. (2) multiplied by $(1 + \eta_{\text{dis}})/\eta_{\text{dis}}$ from Eq. (1) to get

$$U_m\frac{\mathrm{d}\left([O_2(^1\Delta)]_j + [I^*]_j\right)h_j}{\mathrm{d}x} = U_m[O_2(^1\Delta)]_m\frac{\mathrm{d}h_j}{\mathrm{d}x} - nI_2\frac{1+\eta_{\text{dis}}}{\eta_{\text{dis}}}\frac{\mathrm{d}F}{\mathrm{d}x} - h_j\left(k_8[H_2O]_j + k_9[O_2(^1\Delta)]_j\right)[I^*]_j \tag{13}$$

Integrating Eq. (13) and taking into account Eq. (4) for $[I^*]_j$ and Eq. (12) for N yields

$$N = \left(1 + \frac{1}{\eta_{\text{dis}}}\right) + \frac{k_8\chi + k_9 Y_m}{U_m}\frac{[O_2]_m 2K_e Y_m}{(K_e - 1)Y_m + 1}\frac{\int_0^x F\,\mathrm{d}x}{F} + 2\frac{K_e Y_m}{(K_e - 1)Y_m + 1} \tag{14}$$

The spatial dependence of F is found as[5]

$$F = 1 - \frac{\exp(-\sqrt{x/l_{\text{dis}}})}{[f + \exp(-\sqrt{x/l_{\text{dis}}})]} \tag{15}$$

where

$$l_{\text{dis}} = \frac{U_m^3 Dc^2}{(2k_3\eta_{\text{dis}} nI_2)^2}\left[\frac{(K_e - 1)Y_m + 1}{K_e Y_m}\right]^2 \tag{16}$$

is the length of the dissociation region and

$$f = \frac{Dc^2 U_m^2 k_2 [O_2]_m Y_m}{2(k_3\eta_{\text{dis}} nI_2)^2}\left[\frac{(K_e - 1)Y_m + 1}{K_e Y_m}\right]^2 \tag{17}$$

Substituting F from Eq. (15) into Eq. (14) yields the final expression for N:

$$N = \left(1 + \frac{1}{\eta_{\text{dis}}}\right) - \frac{k_8\chi + k_9 Y_m}{\eta_{\text{dis}} k_3 \beta}\frac{\ln(1-F)}{F}\frac{h_j(x_{\text{ind}})}{H} + 2\frac{K_e Y_m}{(K_e - 1)Y_m + 1} \tag{18}$$

where $x_{\text{ind}} = l_{\text{dis}} \ln^2(1/f)$ is the induction interval,[5] i.e., the distance between the injection point and the region of rapid dissociation. Comparison between Eq. (9) and Eq. (18) obtained in the cases of premixed flow and finite mixing rate, respectively, shows that the second term of Eq. (18) corresponding to the quenching of electronic excitation in reactions 8 and 9 is $h_j(x_{\text{ind}})/H$ times that of Eq. (9), the other terms being identical. Therefore, formally setting $h_j = H$ in Eq. (18), one gets Eq. (9), which shows that Eq. (18) is more general than Eq. (9).

It is seen that N depends on the dissociation fraction F, the yield Y_m, the titration ratio β, and H_2O and He ratios, χ and ψ, respectively. Figure 2 shows the F dependence of N, for typical parameters of the supersonic COIL. It is seen that for low F the parameter N is almost independent of F, whereas for $F \to 1$, N increases $\propto \ln[1/(1-F)]$. The increase in N is attributable to the increase of the quenching of $O_2(^1\Delta)$ and I^* in reactions 8 and 9. Therefore, to minimize energy losses in the COIL, the dissociation fraction F before the resonator should not be too close to 1. Figure 3 shows the dependencies of N on Y and β. The dependence of Y on N is very weak and nonmonotonic: as Y increases, N decreases for small Y and increases for large Y. The minimal N is achieved at $Y \sim 30\%$. It

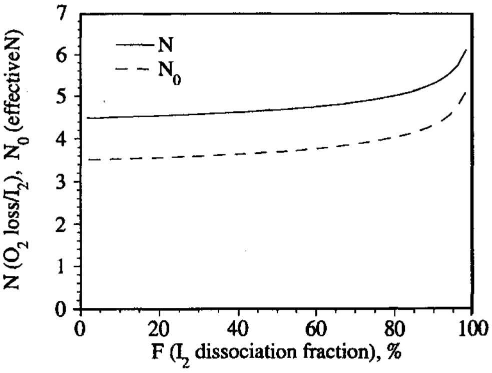


Fig. 2 Dependence of $N(O_2\ \mathrm{loss}/I_2)$ and N_0 (effective N) on the dissociation fraction F. (Values of operation parameters are $Y = 0.4, \beta = 1.5\%, \chi = 10\%, \psi = 3, T = 180$ K, and $h_j = H$.)

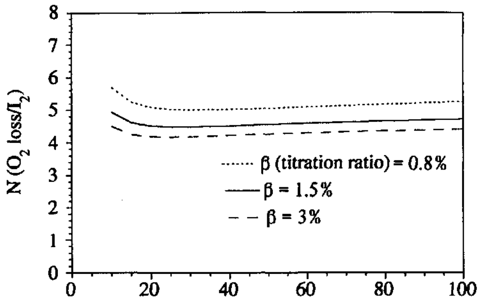


Fig. 3 Dependence of N on the yield Y for different values of the titration ratio β. The values of the operation parameters are the same as in Fig. 2 (F = 0.1).

is seen that N increases as β decreases. Increase in χ and ψ results in the increase of N.

As mentioned above, the third term in Eq. (9) or (18) for N corresponds to the losses for excitation in reaction 1 (Table 1) of I atoms produced via the iodine dissociation. However, part of the energy of the excited I* atoms is converted to the laser energy inside the resonator and therefore is not lost. Indeed, in the case of a Fabry–Perot resonator,[5] the flow rate of I* is given by[5]

$$[\mathrm{I}^*]_{j,\mathrm{res}} = \frac{n\mathrm{I}_2}{h_j U_m} F \frac{2}{3}\left(1 + \frac{g_{\mathrm{th}} H U_m}{\sigma n\mathrm{I}_2 F}\right) \tag{19}$$

where $\sigma = 7.5 \times 10^{-18}(300/T)^{1/2}$ cm^2 (Ref. 5). Comparison between Eqs. (19) and (4) shows that $[\mathrm{I}^*]_j$ before the resonator is greater than $[\mathrm{I}^*]_{j,\mathrm{res}}$. Therefore, the third term in Eqs. (9) and (18) should be replaced with $(\frac{2}{3})[1 + g_{\mathrm{th}} H U_m/(\sigma n\mathrm{I}_2 F)]$. As a result, the effective number N_1, which should be used to calculate the power extracted by a Fabry–Perot resonator, is given by

$$N_1 = N_0 + \frac{2}{3}\frac{g_{\mathrm{th}} H U_m}{\sigma n\mathrm{I}_2 F} \tag{20}$$

where N_0 is the value of N_1 in an ideal resonator with $g_{\mathrm{th}} = 0$:

$$N_0 = \left(1 + \frac{1}{\eta_{\mathrm{dis}}}\right) - \frac{k_8\chi + k_9 Y_m}{\eta_{\mathrm{dis}} k_3 \beta}\frac{\ln(1 - F)}{F}\frac{h_j(x_{\mathrm{ind}})}{H} + \frac{2}{3} \tag{21}$$

Both N_1 and N_0 are smaller than N. Figure 2 shows a comparison between N and N_0. It is seen that $N_0 \sim N - 1$.

III. Influence of $O_2(^1\Delta)$ Losses in the I_2 Dissociation Region on Power

Losses of $O_2(^1\Delta)$ in the I_2 dissociation region strongly affect the dependence of the power on $n\mathrm{I}_2$. Following previous work,[5] it is possible to derive a simple analytical formula for the power:

$$W_1 = 91(\mathrm{kJ/mole}) nO_2 \eta_{\mathrm{mix}}(Y_m - Y_{\mathrm{dis}} - Y_{\mathrm{th}}) \tag{22}$$

where $\eta_{\mathrm{mix}} \equiv h_j(x_{\mathrm{res}})/H$. Taking into account Eq. (12), Y_{dis} is given by

$$Y_{\mathrm{dis}} = N_1 F\beta/\eta_{\mathrm{mix}} \tag{23}$$

$$Y_{\mathrm{th}} = \frac{F + 2\varepsilon}{(2K_e + 1)\{F - \varepsilon[2(K_e - 1)/(2K_e + 1)]\}} \tag{24}$$

is the threshold yield in a Fabry–Perot resonator[5] and $\varepsilon = g_{\mathrm{th}} H U_m/(2\sigma n\mathrm{I}_2)$ is the ratio between the threshold gain and the maximum possible small signal gain. Note that, in Eq. (23) for Y_{dis}, the effective number N_1 is used rather than N.

The I* quenching reactions 3, 8, and 9 of Table 1 are not taken into account in Eq. (22) because the rate of those reactions is much less than the rate of the iodine pumping by reaction 1. Also, the changes of the dissociation fraction F in the resonator are neglected. Equation (22) is similar to the analytical expression for the power derived by the heuristic method of Hon et al.[1] The difference is that the threshold yield Y_{th} given by Eq. (24) is greater than $1/(2K_e + 1)$, the value of Y_{th} employed by Hon et al.[1] [$Y_{\mathrm{th}} \to 1/(2K_e + 1)$ as $\varepsilon \to 0$]. In addition, the number N_1, determining the loss of $O_2(^1\Delta)$, is expressed here as a function of the flow parameters and rate constants, whereas in Hon et al.[1] the empirical value of N was used.

Substituting N_1 from Eq. (20) into Eq. (24), we obtain a final expression for the power:

$$W_1 = 91\left(\frac{\mathrm{kJ}}{\mathrm{mole}}\right) nO_2 \eta_{\mathrm{mix}}\left\{Y_m - \frac{N_0\beta}{\eta_{\mathrm{mix}}}F - \frac{4}{3}\frac{\varepsilon\beta}{\eta_{\mathrm{mix}}} - \frac{F + 2\varepsilon}{(2K_e + 1)\{F - \varepsilon[2(K_e - 1)/(2K_e + 1)]\}}\right\} \tag{25}$$

Because N_0, β, and F are functions of $n\mathrm{I}_2$ [N_0 and F are given by Eqs. (20) and (15–17), respectively], the power W_1 strongly depends on $n\mathrm{I}_2$. Figure 4 shows the $W_1(n\mathrm{I}_2)$ and $F(n\mathrm{I}_2)$ dependencies for the flow conditions close to those of the RADICL[6,18]: $n\mathrm{Cl}_2 = 0.5$ mole/s, $nO_2 = 0.92 n\mathrm{Cl}_2$, $\beta = 1.5\%$, $\chi = 10\%$, $\psi = 3$, the pressure $p = 6$ torr, $U_m = 800$ m/s, $T = 180$ K, $H = 3$ cm, $x_{\mathrm{res}} = 12$ cm, and the channel width $w = 25.4$ cm. The value of Y_m is not known exactly, the estimate, $Y_m = 0.4$, was taken from Carroll,[18] but we also consider a higher values of Y_m in Sec. V. With the above values of parameters, the dissociation efficiency η_{dis} appearing in Eq. (20) for N_0 is 0.78. The dependencies are presented for two values of g_{th}: 0.25 m^{-1} (see Ref. 18) and 0.06 m^{-1}. The first value of g_{th} corresponds to the RADICL mirrors with reflections of the outcoupling and reflecting mirrors of 91 and 99.5%, respectively, absorption/scattering losses of 1% per each mirror and difraction losses of 1% (Ref. 19); also, intracavity losses of 1% are assumed in the present paper.

A simple estimate shows that a large value of the mixing efficiency, $\eta_{\mathrm{mix}} = 0.8$, should be used in the calculations. Actually, in the resonator the dissociation fraction F should be ~1. Taking into account Eqs. (15–17), it is easy to estimate the value of η_{mix} as

$$\eta_{\mathrm{mix}} \cong \frac{K_e Y_m}{(K_e - 1)Y_m + 1}\frac{4k_3(x_{\mathrm{res}}/U_m)n\mathrm{I}_2}{U_m w H}\eta_{\mathrm{dis}} \ln^{-1}\left[\frac{F}{f(1 - F)}\right] \tag{26}$$

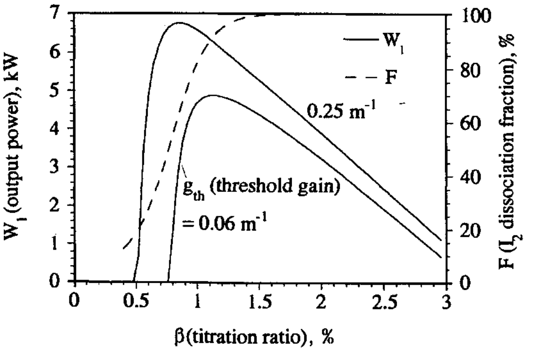


Fig. 4 Dependencies of the power W_1 and dissociation fraction F on $\beta \equiv n\mathrm{I}_2/nO_2$ calculated by Eqs. (25) and (15–17), respectively.

where

$$f = \left\{ \frac{\eta_{\text{mix}} U_m w H}{k_3 n \text{I}_2} \frac{(K_e - 1)Y_m + 1}{K_e Y_m} \right\}^2 k_2[\text{O}_2]_m \frac{Y_m}{2(x_{\text{res}}/U_m)} \tag{27}$$

Substituting the above parameters into Eqs. (26) and (27) and assuming $F = 0.8$, one gets $\eta_{\text{mix}} = 0.8$, i.e., the mixing efficiency is high. The estimated value of η_{mix} is in agreement with the results of the gain measurements in the RADICL reported elsewhere.[20] The width of the gain distribution in the direction perpendicular to the flow velocity measured in that paper makes up for about 80% of the nozzle height at the resonator position. The dependencies of W_1 on the I_2 titration ratio β shown in Fig. 4 have maximums at some optimal values of $\beta = \beta_{\text{max}}$. For $\beta > \beta_{\text{max}}$, W_1 decreases as β increases due to an increase of the losses Y_{dis} in the dissociation region caused by the increase of both iodine molecules, which should be dissociated, and N. The dissociation fraction F_{max} corresponding to the maximum power is always less than one. It is seen in Fig. 4 that the smaller the threshold gain g_{th}, the smaller the value of F_{max}: at $g_{\text{th}} = 0.25 \text{ m}^{-1}$, $F_{\text{max}} = 90\%$, whereas at $g_{\text{th}} = 0.06 \text{ m}^{-1}$, $F_{\text{max}} = 60\%$.

Analytical estimate of F_{max} can be found from Eq. (25), assuming that β changes much less than F:

$$F_{\text{max}} = \left[\frac{6K_e \varepsilon \eta_{\text{mix}}}{(2K_e + 1)^2 N_0 \beta} \right]^{\frac{1}{2}} + \varepsilon \frac{2(K_e - 1)}{2K_e + 1} \tag{28}$$

It is seen that with the decrease of g_{th} and, hence ε, the value of F_{max} also decreases, which is in agreement with the above statement.

It is interesting to compare the iodine flow rate corresponding to the maximum power $(n\text{I}_2)_{\text{max}}$ with the critical flow rate $(n\text{I}_2)_c$ introduced previously[5] and corresponding to the maximum of the gain and the minimum of the iodine dissociation length. As estimated from Eqs. (15–17), $(n\text{I}_2)_{\text{max}}$ is given by

$$(n\text{I}_2)_{\text{max}} \cong \frac{(K_e - 1)Y_m + 1}{K_e Y_m} \times \frac{U_m H}{4k_3 \eta_{\text{dis}}(x_{\text{res}}/U_m)} \eta_{\text{mix}} \ln \left[\frac{F_{\text{max}}}{(1 - F_{\text{max}}) f} \right] \tag{29}$$

whereas[5]

$$(\eta \text{I}_2)_c = \frac{1}{2} U_m \eta_{\text{mix}} H \sqrt{\frac{[\text{O}_2]_m}{k_3 \eta_{\text{dis}}(x_{\text{res}}/U_m)} \frac{[(K_e - 1)Y_m + 1]}{K_e}} \tag{30}$$

The ratio of the preceding flow rates is given by

$$\frac{(n\text{I}_2)_{\text{max}}}{(n\text{I}_2)_c} \approx \frac{1}{2} \sqrt{\frac{1}{k_3 \eta_{\text{dis}}(x_{\text{res}}/U_m)[\text{O}_2]_m}} \ln \left(\frac{1}{f} \right) \tag{31}$$

This value of the ratio is always much less than one, e.g., for the above values of the RADICL parameters, $(n\text{I}_2)_{\text{max}}/(n\text{I}_2)_c \sim 0.3$. Therefore, the iodine flow rate corresponding to the maximum power should be less than that corresponding to the maximum gain.

V. Power and Dissociation Fraction in the RADICL

To check the above analytical expressions, the values of N are calculated numerically for the RADICL, applying the model developed earlier.[5,11–13] In addition, the power and the iodine dissociation fraction as functions of $n\text{I}_2$ are calculated. The nozzle contour and the flow parameters for the RADICL are taken from previous research.[6,18] The results are compared with experimental measurements reported elsewhere.[6] Comparison between the calculated and the measured values of the dissociation fraction and the power makes it possible to estimate the values of the yield Y and the mixing rate.

Figure 5 shows calculated dependencies of W_1 and F before the resonator on $n\text{I}_2$ as compared with experimental results.[6] The empirical entrainment factor c [see Eq. (11)] is equal to 3.2, which is 4 times the value used previously[5]; c was increased because, in the RADICL, the iodine is injected through two rows of the holes, whereas in Ref. 5 only one row is used. As a result, the interfacial area between the jets and the primary flow increases substantially. The mixing efficiency η_{mix}, i.e., the fraction of the main flow occupied by the jets, is about 0.75 at the resonator location, which is in line with the estimate made in Sec. IV [see Eqs. (26) and (27)] as well as other modeling results,[19] where the complete mixing between the jets and the main flow was achieved at the resonator position. The calculations are done for a water ratio $\chi = 10\%$ (Refs. 18 and 19). The value of $Y_m = 35\%$, which gives the best fit to the experimental results, is chosen. The agreement between calculated and measured values of the power and dissociation fraction shown in Fig. 5 is good. The maximum power is achieved when the calculated F is about 80%, which is in agreement with the measurements.[6] For higher Y_m (60%), both dissociation fraction and power rise, as shown in Fig. 5 by dashed curves. For this value of Y_m, agreement between the calculated and experimental values of the power[6] is poor, which is possibly due to higher losses in the optical resonator and hence lower optical extraction efficiency.

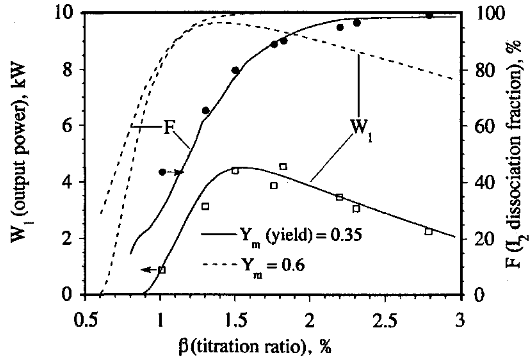


Fig. 5 Calculated and measured[6] power and dissociation fraction in the RADICL ($\psi = 3$, $\chi = \text{H}_2\text{O}/\text{O}_2 = 10\%$, $c = 3.2$).

Calculated values of N do not change strongly and are in the range 4.5–5, which is in agreement with the value obtained from the analytical expression (18), $N = 4.6$.

VI. Conclusions

The preceding considerations show that in the COIL the losses of $\text{O}_2(^1\Delta)$ during the dissociation of the iodine is one of the main factors affecting the power. Analytical expression is obtained for the number of $\text{O}_2(^1\Delta)$ molecules, N, lost in the region of I_2 dissociation per molecule of I_2. This expression yields $N = 4$–6, in agreement with numerical calculations and experimental measurements. It is shown that some effective number $N_1 < N$ should be used rather than N to calculate the power. Analytical expression for the power is obtained taking into account the $\text{O}_2(^1\Delta)$ losses in the dissociation region. It is shown that N increases and the power decreases when the dissociation fraction F increases. Therefore, the maximum power corresponds to low values of $n\text{I}_2$ when iodine is not completely dissociated before the resonator and the small signal gain in the resonator region is not maximal. This statement is in agreement with the results of the RADICL experiments mentioned earlier, where it was found that rapid dissociation did not correspond to the maximum power.

Numerical modeling of the RADICL shows that agreement between calculated and measured power and iodine dissociation fraction can be achieved if the mixing efficiency η_{mix} and $\text{O}_2(^1\Delta)$ yield are about 0.8 and 35%, respectively.

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