PACS numbers: 42.55.Ks; 42.60.Lh DOI: 10.1070/QE2002v032n02ABEH002137

Effect of the solution temperature in a singlet-oxygen generator on the formation of active medium in an ejector oxygen – iodine laser

M.V.Zagidullin, V.D.Nikolaev, N.Yu.Palina, M.I.Svistun, N.A.Khvatov

Abstract. The influence of the solution temperature in a singlet-oxygen generator on the formation of the active medium in the ejector oxygen-iodine laser is investigated. The following parameters of the active medium at the solution temperature $-20\,^{\circ}\mathrm{C}$ are obtained: the gain is $7.2 \times 10^{-3}\,\mathrm{cm}^{-1}$, the Mach number is M=2, the temperature is 205 K, and the static pressure is 9.3 mmHg. As the solution temperature is increased to $-4\,^{\circ}\mathrm{C}$, the gain decreases to $5 \times 10^3\,\mathrm{cm}^{-1}$, the Mach number decreases to 1.78, while the temperature and the static pressure increase to 241 K and 10.7 mmHg, respectively. As the solution temperature increases from $-20\,\mathrm{to}\,-4\,^{\circ}\mathrm{C}$, the losses in $O_2(\Delta)$ increase by less than 20%, while the dissociation efficiency of molecular iodine decreases by less than 21%.

Keywords: singlet-oxygen generator, active medium, oxygen-iodine laser.

1. Introduction

In Ref. [1], the high efficiency ($\sim 20\,\%$) of the ejector oxygen–iodine laser (EOIL) with a high total pressure of the active medium in the cavity was demonstrated. In Ref. [2], the gain, temperature, and absolute velocity of the active medium in the EOIL were determined by the method of diode laser spectroscopy. It was shown that the active medium in the EOIL has a temperature near 200 K, an absolute flow velocity of about 600 m s⁻¹, and a high gain ($\sim 6 \times 10^{-3}$ cm⁻¹). Such a medium possesses a high total pressure, which makes it possible to realise the efficient exhaust of waste gases to the atmosphere.

The formation of the active medium in the EOIL can be described qualitatively as follows [2]. Singlet oxygen (laser energy source) is supplied to the nozzle block from a jet singlet-oxygen generator (SOG) and flows to a mixing chamber through a system of slits with the Mach number M < 1. High-pressure primary nitrogen with an initial

M.V.Zagidullin, V.D.Nikolaev, M.I.Svistun, N.A.Khvatov P.N.Lebedev Physics Institute, Samara Branch, Russian Academy of Sciences, ul. Novo-Sadovaya 221, 443011 Samara, Russia;

N.Yu.Palina Samara State University, ul. akad. Pavlova 1, 443011 Samara, Russia

Received 3 December 2001 Kvantovaya Elektronika 32 (2) 101–106 (2002) Translated by Ram Wadhwa pressure exceeding 1 atm flows into the mixing chamber through a system of cylindrical nozzles and is accelerated over a short distance to M > 2. Between the two flows, a low-pressure flow of nitrogen mixed with molecular iodine is injected.

The expansion of high-pressure nitrogen jets leads to the compression of oxygen jets and the mixture of nitrogen with iodine. The boundaries of downstream primary nitrogen jets form an aerodynamic throat in which the oxygen-iodine mixture flows with the sound velocity. The oxygen-iodine flow expands because the static pressure of this flow in the throat region is higher than the static pressure of the nitrogen flow. As a result, an aerodynamic nozzle is formed, whose walls are the boundaries of the high-pressure nitrogen jets.

Observations show that the oxygen-iodine flow mixes rapidly with the flow of high-pressure nitrogen [1]. The main processes leading to the dissociation of molecular iodine and, hence, to the formation of the active medium, occur in the flow region between the nozzle block and the aerodynamic throat. On this segment, both oxygen and the nitrogen-iodine mixture move at a subsonic velocity under a high pressure. The emergence of atomic iodine in the flow initiates the reactions of deactivation of $O_2(^1\Delta)$. Water molecules, which are always present in a flow of $O_2(^1\Delta)$, actively participate in these reactions. The higher the temperature interval of the solution in the SOG in which the energy and amplification parameters of the active medium change insignificantly, the better the mass and size characteristics of the laser. It was shown in [2] that an increase in the solution temperature leads to a decrease in the gain, but does not affect the rate of its decrease along the flow.

In this work, we study in detail the effect of the solution temperature in the SOG on the formation of the active medium in the EOIL. The singlet-oxygen losses and the efficiency dissociation of iodine molecules as functions of the solution temperature are calculated from the measurements of gas-dynamic parameters of the active medium using the method of laser diode spectroscopy.

2. Measurement and calculation of active medium parameters

The supersonic EOIL is described in detail in Refs [1, 2]. The active medium is formed by an ejector nozzle block to which high-pressure nitrogen, a mixture of nitrogen and iodine vapour, and singlet oxygen $O_2(^1\Delta)$ are supplied. The $O_2(^1\Delta)$ molecules are formed in the jet SOG as a result of the chemical reaction of chlorine with an alkaline solution

of hydrogen peroxide. For the chlorine flow rate $G_0 = 39.2$ mmole s⁻¹, the degree of its utilisation η is 95%, while the yield of $O_2(^1\Delta)$ is $Y_0 \approx 0.6$. In our experiments, the flow rate G_1 of high-pressure nitrogen at temperature $T_1 = 298$ K was 270 mmole s⁻¹, the flow rate G_2 of nitrogen carrying iodine vapour at temperature $T_2 = 333$ K was 11 mmole× s⁻¹, and the flow rate of iodine vapour was $G_{I_2} = 0.8$ mmole s⁻¹.

The flow formed by the nozzle block moved in the mixing chamber with the initial cross section 16×50 mm (the plane angle between the broad walls of the chamber was 4°). We measured the following pressures: p_1 in the SOG, p_2 in front of the oxygen nozzles, p_3 at the wall of the mixing chamber, and p_4 in the Pitot tube. The hole for measuring pressure p_3 was made in the wall of the mixing chamber at a distance of 64 mm from the nozzle block. The Pitot tube was mounted at the centre of the mixing chamber cross section at a distance of 22.5 mm downstream from the hole where the pressure gauge p_3 was installed. In aerodynamic tests of the ejector nozzle block, the chlorine flow was replaced by an air flow with a flow rate of 39.2 mmole s⁻¹ through the SOG. In these tests, the Pitot tube with an outer diameter of 2 mm could be moved along and across the flow in a plane equidistant from the larger walls of the mixing chamber.

equidistant from the larger walls of the mixing chamber. The small-signal gain at the ${}^2P_{1/2}(F=3) \rightarrow {}^2P_{3/2}(F=4)$ transition is given by

$$g(X) = \frac{7}{12} \Delta N \frac{A\lambda^2}{8\pi} \Phi(X), \tag{1}$$

where ΔN is the total inverse population at the $^2P_{1/2} \rightarrow ^2P_{3/2}$ transition; $A=5.1~{\rm s}^{-1}$ is the probability of the $^2P_{1/2}(F=3) \rightarrow ^2P_{3/2}(F=4)$ transition; $\lambda=1.315\times 10^{-4}$ cm is the radiation wavelength; $\Phi(X)$ is the normalised Voigt function depending on the Doppler width $W_{\rm D}$ and on the collision width $W_{\rm L}$ of the transition line; and $X=v-v_0$ is the detuning relative to the line centre. The inverse population in the active oxygen-iodine medium can be represented in the form

$$\Delta N = N_{\rm I} \, \frac{(Y - Y_{\rm th})(K_{\rm eq} + 0.5)}{(K_{\rm eq} - 1)Y + 1},\tag{2}$$

where T is the temperature of the medium; $Y = [O_2(^1\Delta)] \times [O_2]^{-1}$ is the fraction of $O_2(^1\Delta)$ in the active medium; N_I is the concentration of atomic iodine; $K_{\rm eq} = 0.75 \times \exp(401 \, {\rm K}/T)$ is the equilibrium constant of the process

$$O_2(^1\Delta) + I(^2P_{3/2}) \leftrightarrow O_2(^3\Sigma) + I(^2P_{1/2}),$$
 (3)

and $Y_{\rm th}=(2K_{\rm eq}+1)^{-1}$ is the threshold fraction of ${\rm O_2}(^1\!\varDelta)$ for which $\Delta N=0$.

The scheme of measurements of the gain spectrum of the active medium by the method of high-resolution laser diode spectroscopy and its mathematical processing are described in detail in Ref. [2]. The gain spectrum of the active oxygen-iodine medium was recorded using a high-resolution laser diode setup (Physical Science) placed at our disposal by the US Air Force Research Laboratory [3]. The beam emitted by a diode laser passed through the active medium at an angle $\varphi=27.5\,^{\circ}$ to the normal to the velocity vector of the active medium at the centre of the mixing chamber and was at a distance of 64 mm from the nozzle block. As a result, the complete gain spectrum of the active medium for

this beam was the sum of two functions (1) displaced relative to each other by $2\Delta v = 2(\sin \varphi)U/\lambda$ due to the Doppler effect, where U is the flow velocity. The Doppler (W_D) and the collision (W_L) widths of the transition line were determined from the shape of the spectrum g(X).

We determined the following parameters of the active medium: the gain g(0) at the line centre averaged over the path length of the diode-laser beam; the temperature

$$T = (W_D/14.49 \text{ MHz K}^{-1.2})^2,$$
 (4)

the flow velocity

$$U = \frac{\lambda \Delta v}{\sin \varphi},\tag{5}$$

the Mach number

$$M = U \left(\frac{\mu}{kRT}\right)^{1/2} = 14.49 \frac{\lambda \Delta v}{W_D \sin \varphi} \left(\frac{\mu}{kR}\right)^{1/2} \tag{6}$$

(where μ is the average molar weight of the active medium; R is the universal gas constant; and k = 1.4 is the adiabat constant); and the stagnation temperature

$$T^* = T(1 + 0.2M^2) = T + \frac{(k-1)\mu U^2}{2kR}.$$
 (7)

Assuming that the active medium consists mainly of nitrogen and oxygen, we can find the static pressure

$$p = \frac{W_{\rm L}}{c_{\rm N_2} \alpha_{\rm N_2} + c_{\rm O_2} \alpha_{\rm O_2}},\tag{8}$$

where $\alpha_{\rm O_2} = (300~{\rm K/T})^{0.87} \times 5~{\rm MHz~mmHg^{-1}}$ and $\alpha_{\rm N_2} = (300/T~{\rm K})^{0.87} \times 5.5~{\rm MHz~mmHg^{-1}}$ are the collision broadening coefficients; and $c_{\rm O_2}$ and $c_{\rm N_2}$ are the molar fractions of oxygen and nitrogen. The collision broadening coefficients for $T=300~{\rm K}$ are borrowed from Ref. [4] and their temperature dependences are taken from Ref. [5].

We can now find the stagnation pressure

$$p^* = p(1 + 0.2M^2)^{3.5} (9)$$

and the calculated pressure behind the direct shock wave or in the Pitot tube,

$$p_{\rm P} = \frac{166.7M^7}{(7M^2 - 1)^{2.5}} \, p \,. \tag{10}$$

In our calculations, we should make certain assumptions concerning the type of flow mixing. The temperature, velocity, and Mach number calculated by formulas (4)—(6) are valid for the layers in the flow in which the gain takes place or iodine atoms are present. The structure of the flow coming from the nozzle block is close to two-dimensional; namely, the composition and form of the gas flow can be regarded as nearly homogeneous over the height of the nozzle block.

We now assume that we are dealing with complete mixing of the secondary nitrogen flow (containing iodine) with the oxygen flow, while mixing of the oxygen-iodine flow with the primary nitrogen flow is partial. Thus, the gas flow in the mixing chamber is assumed to be two-dimen-

sional, with the layers of pure nitrogen and oxygen—iodine—nitrogen mixture alternating along the nozzles. The velocity U determined by expression (5) is the absolute velocity of the oxygen—iodine layer.

The efficiency of mixing will be characterised by the parameter $\eta_{\rm m}$ indicating the fraction of primary nitrogen mixed with the oxygen-iodine flow. The ratio of the molar fractions of oxygen and nitrogen in such an oxygen-iodine flow is $c_{\rm N_2}/c_{\rm O_2} = (\eta_{\rm m}G_1 + G_2)/G_0$. For $\eta_{\rm m} = 1$, all the three flows mix completely and primary nitrogen does not penetrate into the oxygen-iodine flow for $\eta_{\rm m} = 0$.

Knowing the stagnation temperature of the flow, we can determine the thermal power released in the active medium:

$$Q = T^*(C_0G_0 + C_1\eta_{\rm m}G_1 + C_2G_2 + G_{\rm I}, C_{\rm I_2})$$

$$-(T_0C_0G_0 + T_1C_1\eta_{\rm m}G_1 + T_2C_2G_2 + T_2G_{\rm I},C_{\rm I}), (11)$$

where C_i are specific molar heat capacities of gases; G_i are the molar flow rates of gases; and T_i are the initial temperatures of gases before their mixing in the nozzle block. The temperature T_0 of the oxygen flow in front of the nozzle block was not measured in these experiments. The temperature of oxygen coming from the jets of the solution in the SOG is close to the solution temperature. While calculating the value of Q, we assumed that $T_0 = 273.15 + t$, where t is the solution temperature in degrees Celsius. In fact, this means that while calculating the thermal power, we also take into account the heat release during the transportation of $O_2(^1\Delta)$ from the jets to the nozzle block. The entire thermal power is released in the active medium due to the decrease in the $O_2(^1\Delta)$ content by

$$Y_{\mathbf{q}} = \frac{Q}{q_{\Delta}G_{0}\eta},\tag{12}$$

where $q_{\Delta} = 94.3 \text{ kJ mole}^{-1}$ is the excitation energy of one mole of $O_2(^1\Delta)$. The quantity Q may also contain the heat of condensation of water vapour if it takes place in the cold supersonic flow. The corrections introduced to the estimate of losses in $O_2(^1\Delta)$ due to possible condensation of water vapour will be considered below.

The dissociation of molecular iodine occurs at the expense of the energy of $O_2(^1\Delta)$. In this process, the following fraction of $O_2(^1\Delta)$ is consumed:

$$Y_{\rm d} = \frac{\eta_{\rm d} G_{\rm I_2} q_{\rm I_2}}{\eta G_0 q_{\rm \Delta}},\tag{13}$$

where $\eta_{\rm d}$ is the efficiency of iodine dissociation and $q_{\rm I_2}=148.9~{\rm kJ~mole^{-1}}$ is the dissociation energy of molecular iodine. The content of ${\rm O_2(^1\!A)}$ in the active medium decreases to $Y=Y_0-Y_{\rm q}-Y_{\rm d}$, and the potentially useful fraction of singlet oxygen is $Y_{\rm ext}=Y-Y_{\rm th}$.

The spectrum of the distributed gain g(X) can be used to determine the inverse population averaged over the path of the diode-laser beam:

$$\Delta N = \frac{12}{7} \frac{8\pi}{A\lambda^2} \int_{-\infty}^{\infty} g(X) dX$$

$$= N_{\rm I} \frac{(Y - Y_{\rm th})(K_{\rm eq} + 0.5)}{(K_{\rm eq} - 1)Y + 1}.$$
(14)

Here, $N_{\rm I}$ is the concentration of iodine atoms averaged over the beam path. We can now find the flow rate of atomic iodine in the mixing chamber:

$$G_{\rm I} = 2\eta_{\rm d}G_{\rm I_2} = SUN_{\rm I}$$

$$= SU\Delta N \frac{(K_{\rm eq} - 1)Y + 1}{(Y - Y_{\rm th})(K_{\rm eq} + 0.5)},$$
(15)

where $S = 10 \text{ cm}^2$ is the cross-section area of the mixing chamber at a distance of 64 mm from the nozzle block. Eqns (13)–(15) allow us to estimate the dissociation efficiency of molecular iodine.

3. Results

As the temperature t of the solution in the SOG changes from -20 to -4 °C, the pressures in the SOG and in front of the nozzle block remain unchanged and are equal to $p_1 = 34$ mmHg and $p_2 = 27$ mmHg, respectively, while the pressure p_3 increases monotonically from 8.1 to 9.7 mmHg. For $G_{1_2} = 0$ and t = -16 °C, $p_2 = 26$ mmHg and $p_3 = 6.2$ mmHg, while in aerodynamic tests the values of pressure are $p_2 = 24.7$ mmHg and $p_3 = 5.7$ mmHg.

Figs 1 and 2 show the dependences of the most important parameters of the active medium on the solution temperature in the SOG. The variation of gas-dynamic parameters is in qualitative agreement with their dependence on the heat released during mixing of gas flows in the supersonic ejector [6]. The absolute velocity of the active medium slightly decreases upon heating of the solution in the SOG and amounts to 578 m s⁻¹ for t = -20 °C.

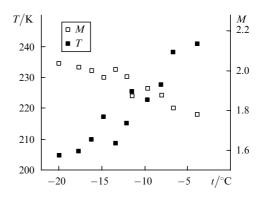


Figure 1. Dependences of the temperature and Mach number of the active medium flow on the solution temperature.

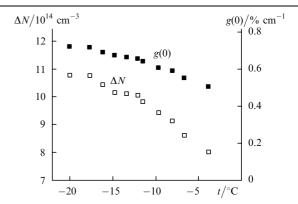


Figure 2. Dependences of the gain and inverse population on the solution temperature.

The collision broadening parameter $W_{\rm L}$ was virtually independent of the solution temperature, but the calculated static pressure increased due to the increase in the temperature of the active medium (Fig. 3). The values of pressure were calculated from the collision width under the assumption of complete mixing of flows ($\eta_{\rm m}=1$). A systematic difference (by approximately 1 mmHg) is observed between the calculated static pressure p and the pressure p_3 at the walls of the mixing chamber. The calculated pressure $p_{\rm P}$ defines the average pressure behind the direct shock wave for oxygen–iodine regions of the flow and slightly differs from the pressure p_4 measured in the central part of the mixing chamber.

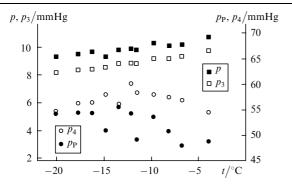


Figure 3. Dependences of pressures on the solution temperature

The distribution of pressure p_4 obtained in aerodynamic tests is presented in Fig. 4. The nonuniformity in the distribution of p_4 across the flow decreases downstream, and the deviation from the average pressure equal to 49 mmHg amounts to approximately 7% at a distance of 90 mm from the nozzle block. Taking into account the inhomogeneity in p_4 in the aerodynamic tests, we can assume that the closeness of the pressure p_P expected behind the direct shock wave front to the pressure p_4 measured in the Pitot tube is quite satisfactory.

The calculated values of pressure slightly increase with decreasing parameter $\eta_{\rm m}$ because the collision broadening coefficient used for nitrogen is higher than the corresponding coefficient for oxygen. The total pressure p^* of the flow

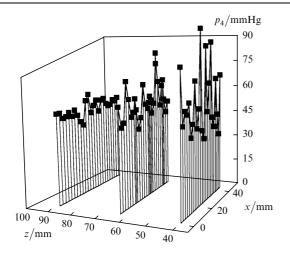


Figure 4. Pressure distribution in a Pitot tube along the flow (z axis) and perpendicular to it (x axis).

regions in which the gain occurs amounts approximately to 70 mmHg, according to calculations, which is almost twice as high as the initial oxygen pressure p_2 . The increase in the total pressure of the oxygen-iodine flow indicates the penetration of high-pressure primary oxygen into the oxygen-iodine flow.

The calculated dependence of the $O_2(^1\Delta)$ losses on the solution temperature is presented in Fig. 5. An increase in the losses Y_q in $O_2(^1\Delta)$ with the solution temperature in the SOG can obviously be explained by the increase in the $O_2(^1\Delta)$ deactivation rate in the flow region from the nozzle block to the aerodynamic nozzle. Indeed, $O_2(^1\Delta)$ transfers energy to an iodine atom in the rapid reaction (3). This is followed by the reaction $I(^2P_{1/2}) + H_2O \rightarrow I(^2P_{3/2}) + H_2O$ of quenching of excited iodine by water vapour (the rate constant of the reaction is $K_{H_2O} = 2 \times 10^{-12}$ cm³ s⁻¹ [7]). Water vapour also effectively quenches the intermediate excited state of molecular iodine as well as the $^1\Sigma$ states of oxygen, which are ultimately formed at the expense of the energy of $O_2(^1\Delta)$ [8].

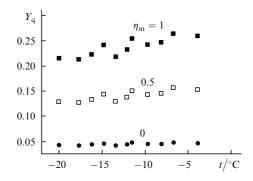


Figure 5. Dependence of the losses in $O_2(^1\Delta)$ on the solution temperature.

As the solution temperature decreases from -4 to $-20\,^{\circ}$ C, the concentration of water vapour in the oxygen flow decreases by several times [9]. Nevertheless, one can see that for any assumed efficiency of mixing $\eta_{\rm m}$, the increase in the value of $Y_{\rm q}$ does not exceed 20 % upon an increase in the solution temperature from -20 to $-4\,^{\circ}$ C. However, the total loss $Y_{\rm q} + Y_{\rm th} + Y_{\rm d}$ of the potentially useful fraction of $O_2(^1\Delta)$ increases at a higher rate upon an increase in the solution temperature because the value of $Y_{\rm th}$ increases additionally.

According to the kinetic models, water vapour should reduce the efficiency of dissociation of molecular iodine [8]. For example, the concentration of electronically excited oxygen $O_2(^{1}\Sigma)$ playing the important role at the initial stage of dissociation of molecular iodine is inversely proportional to the water vapour concentration. The results of calculation of the dissociation efficiency for molecular iodine are presented in Fig. 6. The relative decrease in the value of η_d upon heating of the solution from -20 to -4 °C does not exceed 21 % even for $\eta_m = 0$ despite the fact that the water vapour concentration in the oxygen flow increases in this case by several times.

4. Discussion of results

The calculation of temperatures T and T^* , the absolute velocity, and the Mach number from the gain spectrum are

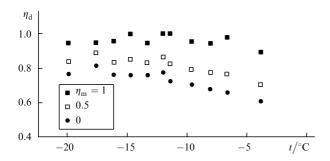


Figure 6. Dependence of the dissociation efficiency of molecular iodine on the solution temperature.

valid only for flow regions containing iodine atoms. The estimates of the $O_2(^1\Delta)$ losses and the iodine dissociation efficiency, as well as the dynamics of their variation upon heating of the solution obtained on the basis of these data depend to a considerable extent on the parameter η_m of mixing of primary nitrogen with oxygen-iodine flow used in our calculations. For example, for $\eta_m = 1$, the losses in $O_2(^1\Delta)$ change most strongly upon heating, while the dissociation efficiency virtually remains unchanged. On the contrary, for $\eta_m = 0$, the calculated losses in $O_2(^1\Delta)$ remain virtually unchanged, while the dissociation efficiency decreases.

Observations reveal that in the case of laser-induced luminescence, the flows mix almost completely at a distance of 64 mm from the nozzle block [1]. On the other hand, in aerodynamic tests, pressure p_4 does not level out over the cross section of the flow even at a distance of 90 mm from the nozzle block. For $\eta_{\rm m}=1$, we have $Y_{\rm q}=0.223,\ Y_{\rm th}=$ 0.09, $Y_{\rm d} = 0.033$, and $Y_{\rm ext} = 0.254$, while for $\eta_{\rm m} = 0.5$, we obtain $Y_{\rm q} = 0.13$, $Y_{\rm th} = 0.09$, $Y_{\rm d} = 0.03$, and $Y_{\rm ext} = 0.38$ at the solution temperature t = -16 °C. For the typical efficiency of transformation of the stored energy into radiation of about 50 % in an EOIL with a gain length of 5 cm [1], the chemical efficiency is 12.5% and 19% for $\eta_{\rm m} = 1$ and 0.5, respectively. The value 19 % is close to that obtained in the experiments [1]. Thus, the uniform mixing of the flows observed in Ref. [1] by using laser-induced luminescence apparently does not indicate their complete mixing up to molecular level.

The decrease in the gain from 7.2×10^{-3} to 5×10^{-3} cm⁻¹ upon heating of the solution from -20 to -4 °C is caused by the following four reasons: the increase in the $O_2(^1\Delta)$ loss, the increase in the Doppler width of the gain line, the increase in the threshold fraction $Y_{\rm th}$, and the decrease in the molecular iodine dissociation efficiency. It would be interesting to estimate the contribution from each factor. According to estimates, the increase in the Doppler width leads to a decrease in the values of $\Phi(0)$ and the gain by a factor of 1.06. The contributions from other factors can be determined only for the known value of $\eta_{\rm m}$. For $\eta_{\rm m}=0.5$, the concentration of iodine atoms decreases by a factor of 1.17, and the decrease in the value of Y and the increase in the threshold fraction $Y_{\rm th}$ result in a decrease in the gain by a factor of 1.16.

The $O_2(^1\Delta)$ losses were estimated by neglecting the heat of condensation of water vapour carried away by the oxygen flow from the SOG. The water vapour pressure p_{s1} in SOG is close to saturation and is equal approximately to 1 mmHg at a temperature -20 °C of the alkaline solution of hydrogen

peroxide [9]. For the oxygen pressure $p_1 = 34$ mmHg in the SOG, the flow rate of water vapour from the SOG is 1.2 mmole s⁻¹, while the partial pressure of water vapour in the mixing chamber is $P_{\rm H_2O} = pG_{\rm H_2O}/G = 0.03$ mmHg. At a temperature of 205 K, the saturated water vapour pressure is $p_{\rm s2} \approx 0.0026$ mmHg.

Thus, necessary conditions exist for water vapour condensation. The thermal power released upon complete condensation of water vapour to saturation equilibrium is $Q_c = q_{H,O}G_{H,O}(1 - p_{s2}/p_{H,O}) = 61 \text{ W}$, where $q_{H,O} = 56$ kJ mole⁻¹ is the heat of condensation of water vapour at T = 205 K. The neglect of this thermal power increases the estimate of losses in $O_2(^1\Delta)$ by $\Delta Y = Q_c(\eta q_{\Delta}G_0)^{-1} = 0.016$. This correction is much smaller than the total losses $O_2(^1\Delta)$. The flow rate of water vapour and the partial pressure $p_{H_{20}}$ increase with temperature, while the temperature of the active medium and the saturation pressure p_{s2} also increase simultaneously. Estimates show that an increase in the solution temperature does not increase the thermal power released during the condensation of water vapour, and their partial pressure in the active medium is even lower than the condensation point at solution temperatures below -8 °C.

The relatively high losses in $O_2(^1\Delta)$ at the solution temperature $t=-20\,^{\circ}\mathrm{C}$ may be due to two reasons. First, molecular iodine plays a more significant role in the deactivation of $O_2(^1\Delta)$ than the water vapour in the flow region where active processes of iodine dissociation and quenching of electron energy take place. The rate of deactivation of $O_2(^1\Delta)$ under conditions of almost instantaneous energy transfer (3) is determined by the quenching $I(^2P_{1/2})$. The quenching $I(^2P_{1/2}) + I_2 \rightarrow I(^2P_{3/2}) + I_2$ is characterised by the rate constant $K_{I_2} = 3.6 \times 10^{-11}$ cm³ s⁻¹ [10]. The ratio of rates of deactivation $I(^2P_{1/2})$ by the water vapour and molecular iodine is $K_{H_2O}G_{H_2O}(K_{I_2}G_{I_2})^{-1} = 0.08$ at $t = -20\,^{\circ}\mathrm{C}$. Thus, at the intermediate stage of dissociation, the quenching $I(^2P_{1/2})$ by molecular iodine dominates

This is confirmed indirectly by the higher values of pressures p_2 and p_3 for $G_{I_2} = 0.8$ mmole s⁻¹ than for $G_{I_2} = 0$. The second reason for the high initial value of Y_q may be the difference between the oxygen temperature T_0 and the solution temperature. The quenching of $O_2(^1\Delta)$ at the segment between SOG and the nozzle block results in an increase in the oxygen temperature and must lead to an elevation of the pressure p_2 in front of the nozzle block. This is confirmed by a higher value of pressure p_2 in actual experiments than in aerodynamic tests with air. The losses in $O_2(^1\Delta)$ can be decreased by choosing a construction of the nozzle block for which the partial pressure of oxygen decreases during its transportation from SOG, and an optimal flow time for the oxygen-iodine mixture from the nozzle block to the aerodynamic nozzle throat is attained.

The calculation of the static pressure is based on the assumption of a complete mixing of flows, as well as on the specific values of the collision broadening coefficient at $T=300~\rm K$ and their temperature dependences. For the other known value $\alpha_{\rm N_2}=6.2~\rm MHz~\rm mmHg^{-1}$ of the broadening coefficient in the presence of nitrogen at $T=300~\rm K$ [11], the theoretical value of pressure p_3 almost coincides with the pressure p_3 at the walls. Thus, the difference between p and p_3 may be due only to the indeterminacy in the values of the collision broadening coefficients or their temperature dependence. Another reason for the difference in the values

of these quantities may be oblique shock waves in the mixing chamber. Despite this, it is obvious that the shape of the gain line provides a quite correct estimate of the main gas-dynamic parameters of the active medium.

5. Conclusions

Thus, our investigations have shown that an increase in the solution temperature in SOG leads to a decrease in the gain and the stored energy, as well as to a change in the gasdynamic parameters of the active medium of the EOIL. At the solution temperature -20 °C, the value of the gain g(0)was 7.2×10^{-3} cm⁻¹, the Mach number was M = 2, the active medium temperature was 205 K, and the static pressure was 9.3 mmHg. An increase in the solution temperature to -4 °C resulted in a decrease in the value of g(0) to 5×10^{-3} cm⁻¹, and the Mach number to 1.78, while the temperature and the static pressure increased to 241 K and 10.7 mmHg, respectively. As the solution temperature increases from -20 to $-4\,^{\circ}\text{C}$, the losses in $O_2(^1\Delta)$ increase by no more than 20 % relative to the total losses, and the dissociation efficiency of molecular iodine decreases by no more than 21 %. The static pressure and the pressure behind the direct shock wave front, calculated from the gain spectrum broadening, are in good agreement with the measured values.

Acknowledgements. This work was supported by the European office of Aerospace Research and Development (EOARD) (Project No. 007016) and supported administratively by the International Science and Technology Centre (Grant No. 1862P).

References

- Zagidullin M.V., Nikolaev V.D., Svistun M.I., Khvatov N.A., Hager G.D., Madden T.J. Kvantovaya Elektron., 31, 30 (2001) [Quantum Electron., 31, 30 (2001)].
- Zagidullin M.V., Nikolaev V.D., Svistun M.I., Khvatov N.A., Anderson B.T., Tate R.F., Hager G.D. Kvantovaya Elektron., 31, 678 (2001) [Quantum Electron., 31, 678 (2001)].
- Tate R.F., Hunt B.S., Helms C.A., Truesdell K.A., Hager G.D. IEEE J. Quantum Electron., 31, 1632 (1995).
- Davis S.J., Kessler W.J., Bachmann M. Proc. SPIE Int. Soc. Opt. Eng., 3612, 157 (1999).
- Zagidullin M.V., Nikolaev V.D., Svistun M.I., Khvatov N.A. Kvantovaya Elektron., 31, 373 (2001) [Quantum Electron., 31, 373 (2001)].
- Abramovich G.N. Prikladnaya gazovaya dinamika (Applied Gas Dynamics) (Moscow: Nauka, 1976).
- 7. Burde D.H., McFarlane R.A. J. Chem. Phys., 64, 1850 (1976).
- 8. Komissarov A.V., Goncharov V., Heaven M.C. *Proc. SPIE Int. Soc. Opt. Eng.*, **4184**, 7 (2001).
- Furman D., Barmashenko B.D., Rosenwaks S. IEEE J. Quantum Electron., 4, 547 (1999).
- 10. Burrows M.D. J. Chem. Phys., 81, 3546 (1984).
- 11. Padrick T.D., Palmer R.E. J. Chem. Phys., 62, 350 (1975).