Quantum Electronics **33** (12) 1038 – 1042 (2003)

LASERS

PACS numbers: 42.55.Ks; 42.60.Lh
DOI: 10.1070/QE2003v033n12ABEH002550

A continuous-wave DF chemical laser on an ethylene-based fuel composition

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Abstract. **The consequences of replacing the primary fuel, hydrogen, as a part of the conventional $F_2 - H_2 - He$ DF-laser fuel composition by ethylene are studied experimentally. During comparative tests of the laser with different primary fuels, the completeness of combustion of the fuel mixtures is assessed and their optimal chemical compositions, at which the energy and spectral characteristics of laser radiation are measured, are determined. It is shown that, when C_2H_4 is used instead of H_2, an increase in the specific laser energy yield of 37 % without a change in the active-medium length is ensured and radiation within the range of 3.75 – 4.18 μm is generated.**

Keywords: *chemical laser, optimisation of parameters, active media.*

1. Introduction

A cw DF chemical laser has been long known [1]. An interest in this laser is primarily determined by a good transmission of its radiation ($\lambda \sim 3.8$ μm) through the atmosphere along near-surface horizontal and vertical paths [2]. It is precisely this factor that was decisive in the development of high-power location and other ground-based systems using this laser [3].

In the development of such systems, the problem of choosing the fuel composition for the atomic-fluorine generator is of great importance. The composition must provide a combination of high laser energy characteristics with acceptable performance features of the fuel components. In the existing foreign DF-laser-based systems, the $NF_3 - C_2H_4 - He$ fuel is used. Its components are nitrogen trifluoride (NF_3), ethylene (C_2H_4), and helium (He). This may testify to the fact that this fuel satisfies the above requirements to the highest degree. However, no motivated justification of using this fuel is available in the literature.

Domestic experimental studies in the field of DF lasers were performed using only a conventional (basic) $F_2 - H_2 - He$ fuel and were rather limited, since deuterium (D_2), which serves as the secondary fuel with a comparatively high mass flow rate, is very expensive. There is an experience of working only with nitrogen trifluoride used as a fluorine-containing compound in tests of HF lasers based on the $NF_3 - D_2 - He$ fuel mixture.

This work is devoted to the experimental assessment of the consequences of replacing the primary fuel – hydrogen (H_2) – as part of the $F_2 - H_2 - He$ conventional DF-laser fuel composition by ethylene. The necessity of formulating this problem is determined by the potential performance and energy advantages of using ethylene, which allows us to believe in an increased efficiency of the DF-laser operation.

The performance advantages of ethylene can be evaluated by comparing its most important properties to those of hydrogen. The physical properties of ethylene allow it to be stored in the liquid state at normal temperature and a pressure of ~ 5 MPa, which is very important from the viewpoint of reducing the dimensions of the laser facility. The chemical properties of ethylene characterise it as an ecologically safe substance, since the probability of an explosion of its mixture with air is much lower than the corresponding probability for a hydrogen – air mixture. The biological effect of ethylene on a human organism allow it to be classified as belonging to the third (not the highest) class of danger. As to its cost, the advantage of ethylene is insignificant.

The main objective of our study was to experimentally evaluate the energy advantages of the DF laser operating with ethylene.

2. Arrangement of experiments and experimental conditions

We studied the operation an autonomous supersonic DF laser designed to emit ~ 5 kW of output power. The laser has a nozzle assembly with the 25×2.8-cm outlet cross section. The nozzles for supplying the oxidising gas containing F atoms and the secondary fuel (D_2 molecules) are arranged with a spacing of 7.5 mm. The following working reagents were used: gaseous fluorine (the oxidiser), gaseous hydrogen and ethylene (the primary fuel), deuterium (the secondary fuel), and helium (the diluent). The laser was placed in a low-pressure chamber, which was intended for separating from the environment the free-jet gas flow outflowing from the nozzle assembly.

We measured the energy and spectral characteristics of laser radiation and recorded the general pattern of the active-medium flow by its emission in the visible spectral region.

The laser output power was measured by the double-slot-resonator technique [4]. The power measurement system

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Received 19 March 2003; revision received 6 June 2003
Kvantovaya Elektronika **33** (12) 1038 – 1042 (2003)
Translated by A.S. Seferov

consisted of two independent closed resonators, whose optical axes are shifted relative to each other by $\Delta x = x_2 - x_1$ in the direction of the active-medium flow (x_1 and x_2 are the distances from the end of the nozzle assembly to the optical axes of the corresponding resonators). The resonators had rectangularly shaped apertures with a height of 9 cm and widths d_1 and d_2, respectively. The totally reflecting uncooled spherical mirrors with a 5-m radius of curvature were made of polished copper. Each mirror had four chromel – copel thermocouples. The length of the resonators along the optical axis was 1 m. A calcium fluoride plate 16 cm in diameter was used as the exit window of the low-pressure chamber.

The laser power was determined from the temperature increment in each mirror during the lasing time that was specified using a mechanical shutter positioned between the active medium and mirrors. The radiation extracted through a 2-mm aperture in the mirror of the first (in the downstream direction) resonator was directed to a fast MG-30 pyroelectric detector whose signal was recorded by with H-117 oscilloscope and used to measure the lasing duration. The resonators used in the experiments had the following geometrical parameters: $x_1 = 2$ cm, $x_2 = 5.5$ cm, $\Delta x = 3.5$ cm, $d_1 = 4$ cm, and $d_2 = 3$ cm. The laser output power N was determined as the sum of the powers N_1 and N_2 generated by each resonator. The power measurement error of this technique is within $\pm 7\,\%$.

The laser emission spectrum was recorded with an IR spectrometer based on an IKM-1 monochromator, in which the standard dispersive element (a NaCl prism) was replaced by a diffraction grating. An FSG-22-3A2 photoresistor based on a binary Ge – Au compound with the wavelength sensitivity range of 1.8 – 8.5 μm was used as a photodetector. This technique of measuring the relative intensities of the spectral lines ensured an error of within $\pm 5\,\%$. The radiation extracted through the hole in the mirror of the first (in the downstream direction) resonator was used in spectral measurements.

The general pattern of the active-medium flow was recorded using its intrinsic emission in the visible spectral region by a Sony video camera with a film rate of 24 frames per second through the inspection window in the low-pressure chamber.

3. Experimental results and discussion

As was mentioned in Introduction, the main objective of this study was to experimentally assess the results of replacing the primary fuel H_2 in the basic DF-laser fuel composition by ethylene. A convenient technique for this assessment is to compare the characteristics of a laser during its operation with each type of the fuel. To objectively evaluate the laser characteristics, it is obviously necessary to compare them at such chemical fuel compositions that ensure the maximum energy parameter for each of them, for which the specific energy yield $N_{\Sigma}^{\max}$ is usually considered (determined relative to the total mass flow rate of the reagents).

According to this approach, we performed the study in three stages. The first and second stages were devoted to optimising the chemical compositions of the basic and alternative fuels for the DF laser with conventional formulas

$$[H_2 + \alpha F_2 + \psi(\alpha - 1)He] + \alpha_2(\alpha - 1)D_2, \qquad (1)$$

$$[C_2H_4 + 6\alpha F_2 + 6\psi(\alpha - 1)He] + 6\alpha_2(\alpha - 1)D_2, \qquad (2)$$

respectively, where α is the oxidiser-excess factor (the number of fluorine moles per mole of the primary fuel, hydrogen or ethylene); ψ is the degree of fuel-mixture dilution (the number of moles of the diluent, helium, per mole of conventionally molecular fluorine as a component of the combustion products); and α_2 is the factor of the secondary-fuel excess (the number of moles of the secondary fuel, deuterium, per mole of conventionally molecular fluorine). Formula (1) is generally accepted, and formula (2) is written by analogy with (1), thus facilitating the comparison of the results obtained for different fuel compositions. The dimensionless factors $\alpha_{\rm opt}$ and $\psi_{\rm opt}$ were determined during the optimisation. The dimensionless factor α_2 remained fixed and equal to ~ 10 at a fixed pressure $p_c \sim 0.1$ MPa in the combustion chamber of the atomic-fluorine generator.

At the third stage, the energy, spectral, and spatial characteristics of the laser active medium and radiation were measured for lasing with each of the compared fuels under the determined optimal conditions. The characteristics obtained were compared to each other.

3.1 Optimising the chemical compositions of the fuel mixtures

The optimisation has been performed according to the technique proposed in Ref. [5]. Its results are shown in Fig. 1. The two first curves (Figs 1a, 1b) are the functions $N_{\Sigma} = f(\beta)$ plotted for $\alpha = \text{const}$, whose values are taken close to the optimal values obtained in calculations for each fuel (the factor of dilution β of the fuel mixture in the atomic-fluorine generator is the number of helium moles per mole of the primary fuel [5]). With an allowance for (1) and (2) for the laser operation with hydrogen and ethylene, $\beta = \psi(\alpha - 1)$, $\beta = 6\psi(\alpha - 1)$.

Two other dependences (Figs 1c, 1d) are the functions $N_{\Sigma} = f(\alpha)$ plotted for $\beta_{\rm opt} = \text{const}$, whose values are obtained from the two first dependences (Figs 1a, 1b). Processing all these dependences made it possible to determine the optimal chemical fuel compositions charac-

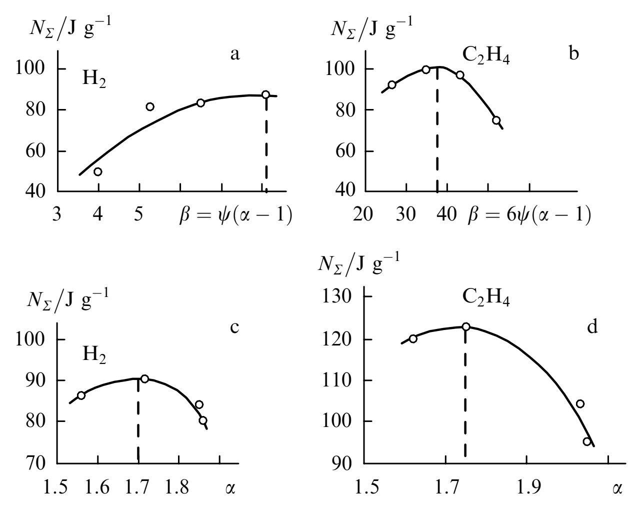


Figure 1. Specific energy yield as a function of (a, b) the degree of dilution β and (c, d) the oxidiser-excess factor α for the DF-laser operation with (a, c) hydrogen and (b, d) ethylene.

terised by dimensionless coefficients α_{opt} and ψ_{opt}. The latter are found from the relations $\psi_{opt} = \beta_{opt}/(\alpha_{opt} - 1)$ and $\psi_{opt} = \beta_{opt}/6(\alpha_{opt} - 1)$ for the versions of lasing using hydrogen and ethylene, respectively. Test experiments using hydrogen and ethylene as fuel components were performed at the experimentally found optimal chemical compositions. The data obtained are listed in Table 1.

Table 1. Optimal chemical compositions of the fuels and energy characteristics of the DF laser.

Fuel in the atomic-fluorine generator	α_{opt}	ψ_{opt}	N_Σ^{max}/ J g^{-1}	N_F^{max}/ J g^{-1}	N^{max}/ kW
$H_2 + \alpha F_2 + \psi(\alpha - 1)He$	1.70	12.0	87	403	3.04
$C_2H_4 + 6\alpha F_2 + 6\psi(\alpha - 1)He$	1.75	8.3	119	505	3.97

We estimated the completeness of combustion of different fuels in one and the same combustion chamber by a conventional method using the pressure-completeness coefficient $\varphi_{p_c} = \mu\varphi_\beta$ [6], where μ is the mixture flow-rate coefficient, $\varphi_\beta = \beta_{exp}/\beta_{theor}$; β_{exp} and β_{theor} are the experimental and theoretical pressure pulses in the combustion chamber. It occurred that the completeness of combustion of various fuels in the chamber 23 cm long is virtually the same and characterised by the coefficient $\varphi_{p_c} = 0.83 - 0.85$.

3.2 Estimating the efficiency of using ethylene

The output energy, spectral, and spatial characteristics of the DF laser were determined at the found optimal chemical fuel compositions and compared to each other. This helped evaluate the efficiency of using ethylene.

The effect of the chemical fuel composition on the DF-laser output characteristics is determined by several factors, the most important of which are the concentrations of atomic fluorine c_F and hydrogen fluoride c_{HF} that are components of the combustion products, their temperature T_c, and the molecular mass m_c. Figure 2 shows the dependences of these parameters on the oxidiser-excess factor α at a pressure in the combustion chamber $p_c = 0.1$ MPa and a degree of dilution ψ_{opt} that was close to the optimal value for each fuel. These data explain why the $C_2H_4 - F_2 - He$ fuel mixture has advantages over the $H_2 - F_2 - He$ basic

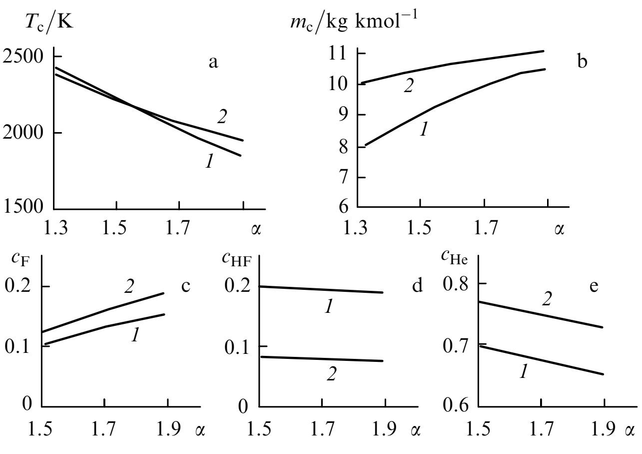


Figure 2. (a) Equilibrium temperature, (b) molecular mass of the combustion products, and equilibrium concentrations of (c) atomic fluorine, (d) hydrogen fluoride, and (e) inert diluent, He, as functions of the oxidiser-excess factor α for the DF-laser operation using (*1*) hydrogen and (*2*) ethylene.

DF-laser mixture from the viewpoint of the output energy (see Table 1).

We see that the use of C_2H_4 instead of H_2 is accompanied by a 20 % increase in the concentration of atomic fluorine in the combustion products of the fuel of optimal composition (Fig. 2c). Since the concentration of atomic fluorine at the end of the oxidation nozzles coincides with its value in the atomic-fluorine generator to a high accuracy (since, according to calculations [7], the flow in the nozzles is virtually 'frozen' in all ranges of changes in the factors α and ψ considered), this leads to an increase in the number of active centres initiating the pump reactions and to the production of additional working DF(v) molecules (v is the vibrational quantum number). A simultaneous 2.5-fold decrease in the concentration of HF(0) molecules, which are the strongest deactivators of the working molecules, takes place and eventually ensures an increase in the laser output power N of 30 %, in the specific energy yield N_Σ of 37 %, and in the specific power per mass flow rate of free fluorine N_F of 25 % (Table 1).

In the light of the experimental data obtained, it becomes clear for what reasons the $C_2H_4 - NF_3 - He$ fuel composition was chosen for the DF-laser-based ground-based systems existing in foreign countries. Nitrogen trifluorine is a less efficient oxidiser than molecular fluorine. Our earlier experimental studies of the consequences of replacing molecular fluorine by nitrogen trifluorine have showed a 35 % decrease in the specific energy yield for a cw chemical laser [8]. A 37 % increase in the output power accompanying the replacement of hydrogen by ethylene fully compensates for this reduction. Thus, the specific energy yield of the DF laser operating on the $C_2H_4 - NF_3 - He$ fuel is actually equivalent to its specific energy yield for the operation on the standard $H_2 - F_2 - He$ mixture. In view of suitable performance characteristics of nitrogen trifluorine and ethylene compared to fluorine and hydrogen, the advantage of the $C_2H_4 - NF_3 - He$ fuel composition becomes obvious.

The spectral distributions of the DF-laser output power (relative powers of the emission lines N_J/N proportional to their intensities) are shown in Fig. 3. They include 18 (Fig. 3a) to 22 (Fig. 3b) lines: $P_0(8) - P_0(15)$ in the $1 \rightarrow 0$ band, $P_1(8) - P_1(16)$ in the $2 \rightarrow 1$ band, and $P_2(8) - P_2(14)$ in the $3 \rightarrow 2$ band for $\lambda = 3.679-4.179$ μm. A characteristic feature of the emission spectrum for a laser operating with hydrogen (Fig. 3a) is the presence of the $P_1(10)$ rotational line at 3.876 μm in the $2 \rightarrow 1$ vibrational band. The relative intensity of this line exceeds the intensities of all other lines by a factor of almost two. Sharp differences in the line intensities are smoothed for a laser operating on ethylene (Fig. 3b). This is determined by the appearance of additional transitions, as a result of which the energy is redistributed over a larger number of states, and the spectral power distribution is shifted to larger rotational quantum numbers J. This shift is accompanied by the disappearance of the $P_0(8) - P_0(9)$ lines and the appearance of the $P_0(14) - P_0(15)$, $P_1(14) - P_1(16)$ and $P_2(13) - P_2(14)$ lines. This fact can be explained by analysing the thermodynamic parameters and chemical composition of the combustion products.

It follows from Fig. 2a that the temperatures of the combustion products for both fuels are identical; therefore, when the products outflow from nozzles of identical configurations, the temperatures at their outlets are close

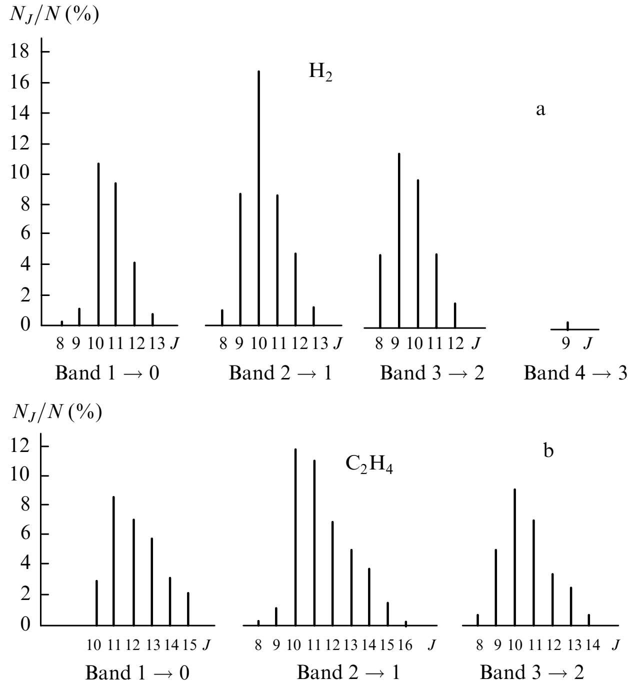


Figure 3. Lasing spectra of the DF laser operating with (a) hydrogen and (b) ethylene.

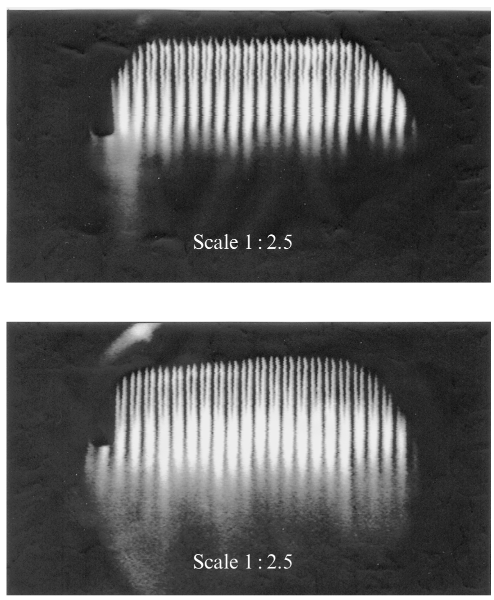
to each other. However, the concentration of fluorine atoms for the laser utilising ethylene is 20 % higher than for the laser operating with hydrogen (Fig. 2c), and the concentrations of the inert diluent (helium) differ by 10 % (Fig. 2e). Therefore, the temperature of the active medium slightly increases due to an increased heat release of the pumping chemical reaction $F + D_2 \rightarrow DF(v) + D$. This favours the formation of the conditions for populating higher rotational levels of the working molecules and finally leads to the red spectral shift. The distribution of the spectral power over bands is characterised by an almost constant ratio between the fractions of the laser output power contained in each band: 26 % in the $1 \rightarrow 0$ band, 43 % in the $2 \rightarrow 1$ band, and 31 % in the $3 \rightarrow 2$ band.

The length of the active medium was estimated from the laser-power distribution over the resonator mirrors and from the data of video filming of the flow. In an optimal lasing mode for each fuel, 79 % and 77 % (21 % and 23 %) of the total laser power fell on the mirrors of the first (second) resonator (in the downstream direction) when using hydrogen and ethylene, respectively. This may testify to approximately identical lengths of the active media, which is confirmed by the results of video filming. As is known [9], the length of the active medium is primarily determined by two parameters: the relaxation losses depending on the chemical composition of the combustion products and the mass flow velocity that depends on the molecular mass of the combustion products and their temperature. The laser operating with ethylene has lower relaxation losses, but the molecular mass of the combustion products is higher compared to that for the laser operating with hydrogen. The mutual compensation for these parameters with an allowance for the close temperatures of the combustion products evidently contributes to the formation of active media of approximately equal lengths. Unfortunately, the available information can hardly help to determine this length. The data obtained earlier for the operation of the model under study in the HF-laser mode on the $F_2 - D_2 - He$ fuel have shown that this length is 7 cm.

The influence of the degree of dilution ψ of the combustion products of the $C_2H_4 - F_2 - He$ fuel with He on the flow structure inside the resonator can be evaluated more definitely. Video filming has shown that, as ψ increases from 4 to 8, the regime of the gas outflow from the oxidation nozzle changes. At $\psi = 4$, the nozzles operate in the incomplete-expansion mode (Fig. 4a); at $\psi > 4$, they operate in a mode close to the designed one (Fig. 4b). In this case, the length of the front, which emits pale-blue light in the flow direction, increases. This may also indicate an increase in the length of the active medium. The dilution has the most pronounced effect on the mixing of jets of the oxidising gas and the secondary fuel. When jets are mixed due to a laminar diffusion, DF-laser radiation is observed within a comparatively narrow layer, which begins in the outlet cross section of the nozzle assembly at the location of a contact between the oxidising-gas and secondary-fuel jets. This reaction zone gradually penetrates into the oxidising-gas jet and reaches its axis in the cross section located at a distance L_{mix} in the downstream direction. The quantity L_{mix} is called the conventional mixing length: this is the distance from the end of the nozzle assembly along the oxidising-gas nozzle axis to the point at which emission is generated on the flow axis [10]. Video frames (Fig. 4) show that, with an increase in the degree of dilution ψ, the L_{mix} value (the length of the dark 'triangular' region against the background of a light oxidising-gas jet) also increases. This is due to a blocking effect of helium on the mutual diffusion of fluorine atoms and deuterium molecules.

4. Conclusions

The experimental study of a cw DF chemical laser has shown that, by replacing hydrogen by ethylene, which serves as the primary fuel in the atomic-fluorine generator,



a

b

Figure 4. Video frames of the active-medium flow in the DF laser that operates using ethylene at a degree of dilution of the fuel mixture by He $\psi = 4$ (a) and 6 (b).

it is possible to improve the lasing efficiency and the conditions for operating the laser facility.

Acknowledgements. The authors thank Yu.L. Samotoev for his help in experiments and B.A. Vinogradov for video filming the process.

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