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TOWARD A COMBUSTION-DRIVEN MIXING GDL

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Résumé. — Nous évaluons l'effet de divers polluants sur le laser thermique à mélange à $N_2/CO_2/He$ de l'ONERA. L'intérêt a porté sur les composés susceptibles d'apparaître dans un système à combustion. Pour nos conditions de chambre (12 bars, 3 000 K), nous trouvons qu'un maximum de 5 % de H_2O , 10 % de H_2 ou 15 % de CO_2 en volume est tolérable. D'autre part, CO a un effet important sur le gain, par suite d'un mauvais couplage vibrationnel avec CO_2 . Si la proportion de CO est grande dans l'écoulement de N_2 , un meilleur effet laser est obtenu avec N_2O qu'avec CO_2 sur notre installation à petite échelle.

Abstract. — The influence of various pollutants on the ONERA $CO_2/N_2/He$ mixing GDL is evaluated. Special attention is paid to those chemicals likely to appear in a combustion-driven system. For our plenum conditions (12 atm, 3 000 K), it is found that at most 5 % H_2O , 10 % H_2 , or 15 % CO_2 by volume can be tolerated among combustion products. It is also observed that CO has a large effect on gain due to lack of coupling to CO_2 . If the CO fraction is large, N_2O proves superior to CO_2 for laser action on the small-scale device used.

Several mixing CO_2 lasers with thermal excitation have been operated in recent years [1-6]. Gains in excess of 4 % cm^{-1} were observed, with laser efficiencies of 1.6 % and specific powers of 25 kW/kg/s [4, 5]. The success of mixing schemes is attributable to several advantages:

(i) the N_2 plenum temperature can be raised to very high values without the penalty of large deactivation by CO_2 and H_2O or He during the expansion;

(ii) because the latter gases are injected cold into the main stream, the static temperature in the mixture downstream is lowered; a small expansion ratio nozzle thus can be used, in spite of the much higher stagnation temperatures;

(iii) a variety of other gas combinations can also be used.

There are, however, a number of difficulties:

(i) the mixing introduces aerodynamic disturbances and flow non-uniformity:

— in the downstream mixing schemes [1, 2], strong shocks are formed due to the different flow velocities; this problem may be cured in a screen nozzle arrangement by allowing the gradients to decay over a distance large compared to the nozzle size [6];

— in the throat mixing scheme [3-5], only weak shocks are formed and the gradients decay in a shorter distance; but the composition of the flow

still varies somewhat from centerline to walls; some refinements can be devised in order to restore excellent uniformity [7];

(ii) axial injectors located on the flow centerline [3-5] are delicate to machine and may be damaged by long cw operation if not cooled properly;

(iii) there is at present no simple, efficient way of producing the hot N_2 with large mass flow rates. Shock tubes [3, 6] and arc heaters [4, 5] have been used so far. Ideally, the system should be powered by a rocket-type burner operating at 2 500 K or more. Unfortunately, most chemical reactions also release large mole fractions of CO_2 , CO , H_2O , and particulate matter.

It is precisely the purpose of the present work to assess the influence of these pollutants on the efficiency, so that guidelines for the selection of appropriate fuel and oxidizer pairs can be established. Various gases thus have been mixed and sent into the arc heater and injector. Our efforts were concentrated on the influence of CO , for which only limited results have so far been reported [6, 8]. Gains were measured, and laser action was obtained from CO_2 and N_2O under different conditions. Some improvements were also made in the injector design.

1. **Set-up.** — The work reported herein was carried out with the set-up described in previous publi-

cations [4, 5]. The 2 MW arc heater can provide a hot flow of gas at a maximum pressure of 12 atm and temperatures ranging from 2000 to 5000 K depending on mass flow rate and electrical power; N_2 , CO, air, H_2 or a combination of any two of these species, can be supplied at the selected mass flow rate through adequate feed lines. The stagnation temperature is adjusted by changing the discharge series resistor; it is best measured, with an accuracy of ± 200 K, by calculating the difference between the electrical power dissipated in the discharge, and the heat carried away by the water cooling the arc assembly. Steady state is reached in 3 seconds, and the runs last usually 5 to 10 seconds altogether.

The nozzle and injector stage is unchanged from its original configuration (Fig. 1), except for the minor

FIG. 1. — Sketch of the supersonic nominal Mach 5 nozzle arrangement and injection schemes studied.

injector change described in the next section, which resulted in an improved flow uniformity. Results obtained by injecting various proportions of CO_2 , N_2O , He, and H_2 will be reported below. Gains have been measured on several rotational lines of CO_2 , and oscillator experiments were performed with a 5-pass multimode cavity made of copper mirrors with a 6 mm thick 5 cm $\times$ 5 cm ZnSe etalon as the output coupler. For this application the etalon is preferred over conventional reflectors because of its well-known transverse mode selecting properties and also because one does not have to use the expensive and fragile partially transmitting dielectric coa-

tings. Due to the Fabry-Perot effect the reflective coefficient can reach 45 % for the resonant cavity modes, although the coefficient at the ZnSe/air interface is only 17 %. Powers were measured with a homemade calorimeter of 0.2 s risetime coupled to differentiating electronics.

2. Injection and flow uniformity. — If energy is to be extracted efficiently, the flow of mixed gases should be as homogeneous and uniform as possible. In the experiments reported in references [4, 5], about 30 % of the N_2 stream near the nozzle walls was left with little or no CO_2 injected, and therefore its vibrational energy could not be extracted efficiently. An improvement thus had to be made in our old injector design. This was done by using larger holes: the gases passing through the holes penetrate deeper into the main hot N_2 stream, thus reaching closer to the nozzle walls; in addition, mixing is completed further downstream in a zone of lower temperature and density, which improves freezing efficiency and minimizes pollutant interference.

The improvement in flow uniformity is seen in figure 2, where 3 injector hole configurations are compared by means of the distribution of small signal gain they achieve about 15 cm downstream

FIG. 2. — Gain distribution for three injector configurations. Injector I: 0.8 mm holes in the nozzle midplane, with 1.3 mm spacing; injector II: same size holes oriented 22° to the midplane, alternately up and down; injector III: same as injector II, but holes are 1.2 mm in diameter and 2.4 mm apart.

from the throat. Note in passing that these distributions are not appreciably different 10 cm further downstream within measurement errors. Small signal gain is only one of the several parameters, like concentration, available laser energy, velocity or temperature, that could be used to probe the flow; it is, however, the easiest to measure. We also verified that the static temperature, as obtained from the gain distribution among the P (*J*) lines, does not increase by more than 30 % from centerline to edges for the new injector.

3. Influence of plenum gas composition. — The potential class of fuels for the combustion-driven GDL is limited. The ideal combustion should give a large fraction of N_2 at temperatures in excess of 2500 K, with minimal concentration of deactivating agents like CO_2 and H_2O . The most attractive candidates are polymers or dehydrogenated organic adducts of the $C \equiv N$ radical, which are, however, toxic, unstable or difficult to prepare for the most part. The best oxidizer is N_2O ; the equivalence ratio ideally should be 2, so that only N_2 and CO but no CO_2 are formed, the CO possessing vibrational relaxation rates of the same order as N_2 . On the other hand, note that the requirements on fuel composition would be much relaxed if some H_2O could be tolerated among the combustion products.

Therefore, we carried out a major effort to determine the influence of CO and H_2O on freezing efficiency. Research on another pollutant, viz. H_2O , and another lasing molecule, viz. N_2O , was also carried out for the sake of completeness. One reservation must be expressed concerning the purity of the products after their heating by the arc. Significant decomposition may occur for some species like O_2 or H_2O . In addition, partial reactions with the evaporated electrode material (Cu vapour) and SiO_2 from the porous walls through which the gases are fed are possible. Appreciable deposits of all these substances on the nozzle walls are observed after several minutes of operation. It does not seem, however, that they have a large effect on freezing efficiency when pure N_2 is used.

4. CO . — The influence of CO on the gain as a function of temperature can be found in figure 3. The net mass flow rate of the $N_2 + CO$ mixtures was kept constant at 100 g/s (3.6 moles/s). The injector was of type III and was fed with a fixed flow rate of $CO_2 + He$ (1.46 and 3.3 moles/s, respectively). The gain on the P_{118} line was measured on the flow centerline. These results establish the following points:

(i) For pure N_2 , the freezing efficiency is maintained from 2500 to 5000 K as seen from the comparison between the solid curve (shaped for a best fit to the data for all mixture compositions) and the dotted line which was obtained numerically taking a constant 80 % figure [4, 5] for the net freezing efficiency (ie including N_2 expansion and CO_2 mixing). The levelling off at about 5000 K mostly reflects the influence of the growing partition function on the population of the 001 level of CO_2 . Croshko *et al.* [7] observed a larger drop beyond 3000 K; this result is obtained if the static temperature in the expanded flow is growing faster, or if a line of lower J is used for the gain measurement.

(ii) Similar trends are seen with $N_2 + CO$ mixtures. The gain, however, is smaller, because the energy defect between CO_2 and CO (224 cm^{-1}) is much larger than between CO_2 and N_2 (18 cm^{-1}). One can

FIG. 3. — Gain vs plenum temperature for various N_2/CO mixtures. The scatter in the data is larger for the 30 and 60 % CO cases because fewer runs were made; meanwhile the arc discharge appeared somewhat less stable.

calculate the gain drop resulting from the mismatch in vibration frequencies; to this end, we use a simple model [9] for two V-V coupled harmonic oscillators in a bath at temperature T with no V-T relaxation, and arrive at:

$$v_1/T_1 - v_2/T_2 = (v_1 - v_2)/T.$$

Here, v_1 and v_2 are the vibration frequencies of the oscillators, and T_1 and T_2 their respective vibrational temperatures. Using this equation and assuming all other parameters equal, we arrive at an approximate gain drop of 40 % if we replace N_2 by CO , assuming $T = 300\text{ K}$ and for a 4000 K stagnation temperature with 100 % freezing efficiency. The gain is actually reduced by nearly 70 %, in satisfactory agreement with reference [8]. This result can be explained by the approximations in the vibrational model, by the fact that the rate constants for V-V transfer from CO are about 10 times smaller than for N_2 at room temperature [10], and also by possible partial decomposition of CO in the arc.

These statements are corroborated by power extraction experiments (Fig. 4). A fixed arc heater mass flow rate of 70 g/s was maintained, the CO mole fraction being varied. The CO_2 and He were kept at 1.2 and 3 moles/s, respectively. The stagnation temperature was held in the range 2500–3000 K, and we estimate the static temperature in the flow at about 350 K. The large drop in laser power with

FIG. 4. — Power and gain results for CO₂ and N₂O; the stagnation temperature is about 3 000 K.

increasing CO fraction results from the inability of the CO₂ to extract the vibrational energy stored in the CO. This effect is enhanced by the degradation of laser cavity performance with low gain gas mixtures: the cavity threshold is reached for gains in excess of 0.75 % cm⁻¹ or thereabouts. The gain is also given for comparison, although it was obtained for 100 g/s flow rate of N₂ + CO. It is anticipated that a larger system (our single-pass gain-length is only 12 cm) with a wider cavity in the flow direction would behave much better, even for the largest CO fractions.

In this respect, it is interesting to examine the results obtained by injecting N₂O instead of CO₂; the 001 level of N₂O lies halfway between those of N₂ (2 331 cm⁻¹) and CO (2 125 cm⁻¹), and fairly efficient coupling with these two molecules is possible. The results are also plotted in figure 4, although some conditions are somewhat different. In particular, the output reflector was a hole coupled copper mirror with 70 % reflection, and the static temperature was lower (estimated 200 K); the lower temperature was obtained with a different injector position at the throat; a flow rate of 1.8 moles/s of N₂ was used. Although the power extracted is lower than for CO₂ with pure N₂, there is almost no dependence vs CO fraction. We believe that these power figures, which to our knowledge are the largest reported to date on a cw N₂O laser, could reach the levels obtained with CO₂. For that, however, it seems desirable to increase the gain length through the flow. Analogous results on comparison of performance were obtained

by Stregack and coworkers [11] with their EDGDL, and the thorough analysis by Biryukov and coworkers tends to the same conclusions [12]. Finally, note that partial oxidation of CO by N₂O in the mixing zone near the injector, where the static temperature is still high, may take place causing loss of gain and laser energy. This combustion is probably negligible with the conditions at which the N₂O laser tests were conducted, but may partly explain why no laser effect was observed under the conditions realized for the CO₂ tests.

5. Pollutants with minor concentrations. — Information has been gathered about H₂O, O₂, CO₂ and H₂. Due to the difficulty of producing a homogeneous mixture of H₂O vapour and N₂ and of avoiding condensation along the lines, we preferred sending appropriate mixtures of air and H₂ through the arc discharge, the H₂ burning to produce H₂O.

Replacing N₂ by an equal flow of air essentially showed that O₂ can be treated as an inert gas, reducing gain in proportion to its mole fraction; a similar result has been reported previously [7]. Adding H₂O caused further reduction in gain and power. The runs were conducted by first waiting for steady state to make a reference measurement, and then sending in H₂ for the final measurement. The ratio of these two values is plotted against H₂O fraction in figure 5. Two sets of data are given: gain as measured on the centerline for injectors type II and III, and power for injector III. It is seen that the bigger holes have a favorable effect, as discussed above. Note that for

FIG. 5. — Relative gain drop vs H₂O fraction; flow rates are: 2.15 moles/s air, 1.46 moles/s CO₂ and 3.16 moles/s He.

these mixtures, the deactivation of N₂ by H₂O is due primarily to the CO₂ which acts as a catalytic agent in the temperature range 300-1 000 K; this is inferred from the corresponding V-T rates [13].

The power, of course, drops faster since the optical cavity becomes less efficient as the gain is reduced. The main conclusion from those curves is that water still has an extremely deleterious effect, although its effect upon the laser efficiency is much less than on conventional GDL's. For atmospheric pressure recovery, the stagnation pressure must be raised to at least 50 atm; then, a 2 % H_2O mole fraction would cause about 50 % loss in gain and available energy. In the absence of H_2O , the freezing efficiency is in the 80-100 % range. If one is to keep the net freezing efficiency above 60 % with the H_2O , then its mole fraction must not exceed 1 %. In addition, note that this result is given for pure N_2 and not for $\text{CO} + \text{N}_2$ mixtures; CO is probably more sensitive to H_2O deactivation [14] than N_2 .

The effect of CO_2 is also interesting. For our tests, the CO_2 was formed by addition of CO to the air sent through the arc heater. Due to the absence of H_2O , the deactivation appears appreciably reduced: typically, for an 8 % CO_2 mole fraction, only 10 % loss of available energy is observed.

The effect of H_2 fraction in a pure heated N_2 stream was briefly examined. An 11 % H_2 fraction caused a 25 % loss in available energy, which is in rough agreement with reference [6]. The result of injection at the throat was carefully studied. Judging from the rate constants in reference [13], H_2 is a potential candidate for lower laser level deactivation; in addition, it has good heat conductivity and can also provide rapid cooling in the throat region. The gain vs H_2 flow rate is presented on figure 6. It is

Fig. 6. — Effect of H_2 injection at the throat on gain; flow rates for N_2 and CO_2 are 2 and 1.46 moles/s respectively.

clear that inversion is indeed achieved, even for the largest H_2 mole fractions. Clearly also, H_2 causes little deactivation if present in the downstream portion, since the gain does not drop appreciably for the largest flow rates (note that in the most

extreme case, the H_2 flow is nearly 2/3 of the total flow). However, the gain remains small, i.e. less than half that obtained with equivalent mole flow rates of He. One may think of the following explanations: (i) the static temperatures are slightly larger than for He mixtures, as actually observed by probing various rotational lines; (ii) the CO_2 lines experience large collisional broadening by H_2 . However, no laser action could be obtained with these mixtures, and we interpret this as indication that H_2 is unable to deactivate the lower level properly.

6. Discussion. — This investigation has established some of the guidelines for the choice of the chemicals most suitable for the combustion-driven mixing GDL, and also has given a more precise idea of potential performance. The main advantages of this type of laser are:

(a) large gains and specific energies: some of the tests done with our system yielded gains in excess of 5 % cm^{-1} (on the P12 line, for the conditions of figure 2 with injector III, but with a larger expansion ratio to give a static temperature of about 150 K). The power exceeds 3 kW for pure N_2 (Fig. 4), the specific power being 50 kW per kg/s of N_2 . We estimate the power wasted because of mirror losses and incomplete saturation to be of the same order. In brief, and including the necessary mass flux of CO_2 and He or H_2O , a large system, properly optimized, should be able to offer specific energies well in excess of 50 kJ/kg, i.e. in the same range as electrical lasers. Recently, three types of mixing arrangements were compared [15], namely injection through the nozzle sidewalls, injection through a central body as on our system, and a screen nozzle of the type developed by Cassady and coworkers [6]. This investigation concluded that the latter was superior, yielding 23 kJ/kg N_2 on a very small device (35 mm active length). Such a comparison should also be made on a larger system, where cavity extraction efficiency depends less on optical gain, for a full evaluation to be made.

(b) The ability to choose the emitted wavelength: operation on transitions of CO_2 and N_2O molecules has been obtained. More molecules, like CS_2 and C_2H_2 , could be made to lase [11], but anticipated efficiencies are lower. Laser action on CO transitions has potential for larger efficiencies than CO_2 , but the drawback is that static temperatures below 150 K must be obtained. We failed to observe CO laser action using hot N_2/CO mixtures and injecting argon. Insufficiently low static flow temperatures, insufficient gain length and large mirror losses are blamed for this negative result.

The effect of some pollutants was assessed. Extremely deleterious effects are seen with H_2O and H_2 , the permissible levels being on the order of 1 % for H_2O , and 2 % for H_2 if the stagnation pressure is to be raised to about 50 atm. Several percent CO_2

are acceptable; this is essential for a combustion driven system, since one wants to achieve high equivalence ratios, as near 2 as possible, but still not have the production of soot which is unavoidable in the vicinity of 2. Strictly speaking, CO cannot be labelled as a pollutant. The available laser energy in the flow is hardly affected by its presence; however, the CO₂ molecule has difficulty in extracting the vibrational energy stored in the levels of CO due to the energy defect. Whether the situation improves much on a larger scale device is still an open question. If not, then N₂O will be an extremely attractive substitute for CO₂ for laser action.

Optical homogeneity has not been determined quantitatively. This will be best done on a system more representative of a large scale laser, i.e. with an array of parallel nozzles offering an optical path of 30 cm or more. A qualitative schlieren study of the present system, in a direction parallel to the throat, had shown [5] that small scale turbulence was of

the same order as that of a conventional GDL flow without injector. We must point out that since the working medium offers more gain and energy, the optical path is reduced for a given output power; this relaxes the requirements on optical quality.

Finally, the production, by combustion, of hot N₂ with an admixture of other acceptable reaction products has yet to be demonstrated. Temperatures of at least 2 500 K and optimally 3 000 K are needed for the laser to be efficient. This restricts the choice of possible fuels considerably. An exhaustive investigation of GDL fuels had been completed by Pallay and Zovko [16]. Few of these fuels appear fully satisfactory for a mixing GDL, either because of excess pollutant formation or because of insufficient combustion enthalpy. The only technically acceptable choice is C₂N₂, which is toxic; preheating may also offer a wider choice of acceptable chemicals. The future of mixing GDL's depends on developments in these areas.

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