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Scaling and Performance of CO_2 Lasers at Supra-Atmospheric Pressure

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Abstract. The performance of a compact uv photo-preionized TE laser is studied in the pressure range 1–5 bar. As the pressure is increased, the laser pulse shape is little altered, but both the peak power and the total output pulse energy increase significantly with pressure, even for constant input electrical energy. For various gas mixtures and excitation source capacitors the measurements suggest approximate output energy scaling with the product of the source charge per unit electrode area [$C. m^{-2}$] and the molecular partial pressure [CO_2+N_2+CO]. This is explained in terms of the pressure-dependent discharge impedance. An input-energy-related discharge instability limits the optimum laser pressure to 1.5–2.5 bar, and we show that, at constant input energy, the instability boundary depends on the molecular partial pressure alone. The pre-ionization photo-electron yield varies negligibly with pressure, but the discharge tolerance to added oxygen decreases as p^{-3} to p^{-4}, dependent on gas mixture. Nevertheless sealed operation for $>10^5$ shots has been obtained with a 5% CO_2:5% CO:3% N_2:2% H_2:85% He gas mixture at a total pressure of 5 bar.

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Most transversely-excited pulsed CO_2 lasers operate at atmospheric pressure. However, this has largely been established because of convenience rather than on the basis of consistent and systematic parametric analysis. TE lasers have been operated occasionally at sub-atmospheric pressures and frequently at higher pressures (up to ~20 atm), but it is difficult to use the published literature to deduce a comprehensive model of pressure scaling since the reports are rather fragmented and unsystematic, and little attention has been paid to explaining the effects of pressure variation on the electron and molecular kinetic (see [1] and references therein).

Frequency tunability of the output of a CO_2 laser is almost continuous for pressures $\gtrsim 10$ bar. In addition, it is often claimed that increasing the gas pressure leads to higher output energies and peak powers or smaller devices for a given output performance, as well as the ability to pump the gas harder before discharge instabilities develop [1]. In this paper we consider these claims, and try to answer the questions:

(i) What is the explanation of improved performance with increased pressure?

(ii) Can the improvement be optimised and what are the correct laser scaling parameters?

(iii) What are the main limitations to sealed laser operation and can long life high pressure devices be realised?

1. Laser Design and Basic Performance

Our experiments used the laser configuration of [2, 3] with the sliding spark uv preioniser array positioned along one side of the main discharge volume and electrically connected in series. The discharged volume is 6 cc between modified Chang profile electrodes in a total gas volume of about 1 l within a high pressure stainless steel chamber. The optical cavity is formed between an internal 4 m gold coated copper full

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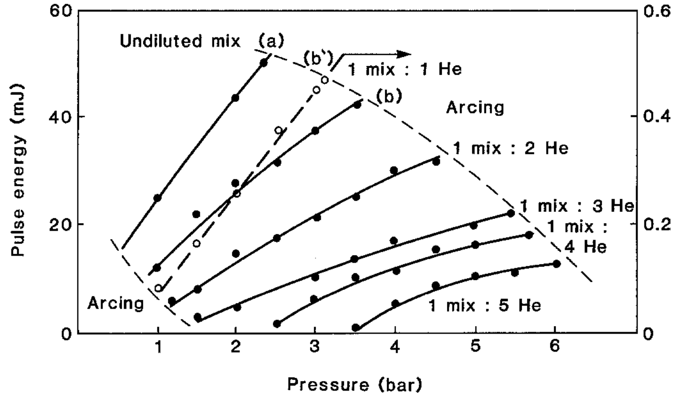

Fig. 1. Measured dependence of laser output energy on total gas pressure for 25% CO_2:15% N_2:15% CO:45% He pre-mixture (Curve *a*), and for increasing He dilutions. Curve *b* shows the pulse energy dependence for a 1:1 dilution ratio (i.e., 12.5% CO_2:7.5% N_2:7.5% CO:72.5% He resultant mixture), and Curve *b*' shows the vatiation of the laser peak power for the same case. (2.7 nF source capacitor, 40 kV charging voltage)

reflector and either an 85% or 95% reflecting flat output coupler mounted in the end flange along with the vacuum and high voltage lead-throughs.

A standard mixture of research grade gases was normally used (25% CO_2, 15% N_2, 15% CO and 45% He) and this was diluted with added helium as required. Results were obtained for stationary sealed mixtures, but the gas was replaced between each set of results to minimise effects due to discharge-induced plasma chemistry (except when such effects were specifically examined as in the long life tests). The laser output was of the normal CO_2 TEA pulse shape with an initial sharp gain-switched spike followed by a long low-amplitude tail. The solid curves of Fig. 1 show the dependence of laser energy output (measured with a Gen-Tec ED 200 Joulemeter) on total gas pressure in the range 1–6 bar, with the charging capacitor C_1 kept constant at 2.7 nF (charging voltage 40 kV). Curve a is for the undiluted pre-mixture and clearly shows an energy output increase with pressure: however the maximum operable pressure is limited to about $2\frac{1}{2}$ atmospheres because of arcing in the main discharge volume. The other curves are for various dilutions of the pre-mixture with helium. As the dilution of the pre-mixture is increased so arc-free operation can take place at higher pressures, but the laser energy is reduced as the amount of the non-helium fraction is reduced. The results of Fig. 1 are for the 15% output coupler; with lesser coupling the output energies were decreased at the lower pressures, but enhanced at higher pressures (in the diluted and hence lower gain mixtures).

The dashed line in Fig. 1 (curve b') shows the variation with pressure of the laser peak power (measured with a Rofin 7441 photon drag detector) for the same mixture as the energy curve b (50% mixture: 50% added helium). The slightly greater increase of peak power with pressure (compared to energy) was typical, and indicated that higher pressures slightly favoured the initial spike rather than the tail of the pulse.

Figure 2a shows the effect of varying the value of the main charging capacitor C_1, for the 6.25% CO_2 mixture (with 5% output coupling, to allow measurements of laser output energy in low gain systems). The output energy increases with both the value of C_1 and the pressure of operation. Now, to assess the effectiveness of the electrical excitation at various pressures it is convenient to use a volume- and -pressure normalising parameter: the parameter "electrical input energy per litre per atmosphere total pressure" [Jl^{-1} atm^{-1}] is often used and has become generally accepted. It is assumed to quantify the amount of excitation of the gas per "average" gas particle. Figure 2b illustrates the

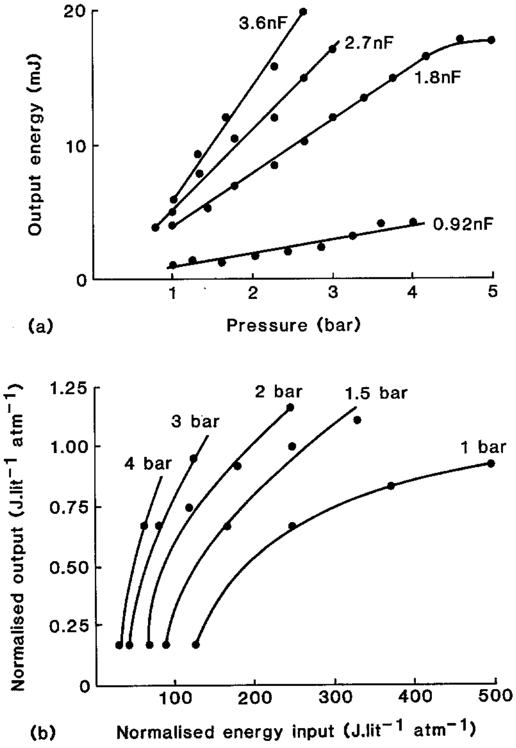

Fig. 2. (a) Effect source capacitance on energy-pressure performance of 6.25% CO_2:3.75% N_2:3.75% CO:86.25% He mixture with 40 kV charging voltage. The results are re-plotted (b) in terms of the customary volume- and pressure-normalising parameter (energy per unit discharge volume per atmosphere total pressure)

results of Fig. 2a re-expressed in terms of this normalised input energy parameter. Clearly "Jl^{-1} atm^{-1}" is not in itself a good indicator of the *output* energy to be expected when the total pressure of the laser gas mixture is varied. Additionally, Fig. 2b displays the saturation of output energy with increasing input energy which results from superelastic electron collisional relaxation of excited molecules (described previously by us [4]).

In the next section we seek appropriate parameters to describe the scaling of output energy with total gas pressure, gas mixture and electrical excitation, and try to explain why the output pulse energy and peak power increase steadily with pressure (Fig. 1) despite the constant electrical energy of the charging capacitor. Sections 3 and 4 examine the effect of pressure changes on the diffuse-discharge/discharge-arcing boundary, and suggest optimum parametric operating areas for arc-free laser output.

2. Pressure Scaling of Laser Output Energy

It is well established in the CO_2 literature that the discharge excitation reduced field E/n becomes constant soon after the breakdown and remains at a "plateau" value whilst most of the current is flowing through the discharge: this suggests an attachment dominated discharge. Recent work by Midorikawa et al. [1] has confirmed the general validity of this model, but suggests there is a small reduction of the plateau value with increasing pressure ($\approx 20\%$ decrease for 2 to 10 atm pressure variation). Such a decrease is not of importance in a preliminary analysis and we will assume E/n constant for a given gas mixture and invariant with current.

The glow discharge impedance r' will be rather low (a few Ω) and usually much less than the impedance of the external circuit R; thus the magnitude of the current will be independent of the gaseous discharge and will be completely controlled by the external circuit. In these circumstances a change of gas pressure should not significantly alter the discharge current i. To check this we used an English Electric MA 391A current transformer and found that although the peak current was not constant, it only decreased by about one third as the pressure was increased from 1 to 5 atm. Furthermore the time-integrated current (charge Q) did remain approximately constant. (The explanation for the concomitant discharge pulse lengthening is complicated and associated with our specific discharge circuit.) In the simplest approximation, then, both the reduced field E/n (or E/p) and the product of average current and excitation pulse length (ixt) $= Q$ are constant with changes of gas pressure p.

The energy deposited in the discharge (per unit volume) is iVt/A where A is the electrode area, $V = El$ is the discharge plateau voltage and l the electrode separation. Using a very simple equivalent circuit [5]

$$W = iVt/A = V(V_0 - V)C/A\,,$$

where the source capacitance C is initially charged to voltage V_0. It is clear that the energy deposited in the gas is not linearly dependent on the energy in the source capacitor ($CV_0^2/2$) and the efficiency of energy transfer is $2V(V_0 - V)V_0^{-2}$. In the regime $r' \ll R$ and $V \ll V_0$ (which are, of course, related) the energy deposited in the gas will be linearly dependent on the source capacitor charge $Q = CV_0$. To elucidate, consider the total vibrational excitation per unit volume integrated over the length of the current pulse:

$$S = \bar{n}_e tp[k_1(E/n)f_{CO_2} + k_2(E/n)f_{CO} + k_3(E/n)f_{N_2}]\,,$$

where f_{CO_2}, f_{N_2}, and f_{CO} are the molecular fractions of CO_2, N_2, and CO in the mixture, p is the total pressure and k_1, k_2, and k_3 are the appropriate mixture-weighted rate constants for vibrational excitation of CO_2 (00_n mode), CO and N_2, respectively. For simplicity we may introduce K, a mixture-weighted rate constant for the total useful vibrational excitation such that

$$S = \bar{n}_e tpK\,.$$

Since E/n is roughly constant with pressure, so too is the drift velocity V_d; the average discharge current is

$$i = \bar{n}_e V_d Ae$$

so we have

$$\begin{aligned} S &= itp\,K/AV_d e \\ &= QpA^{-1} \times (\text{constant}) \\ &= CV_0 pA^{-1} \times (\text{constant}) \end{aligned}$$

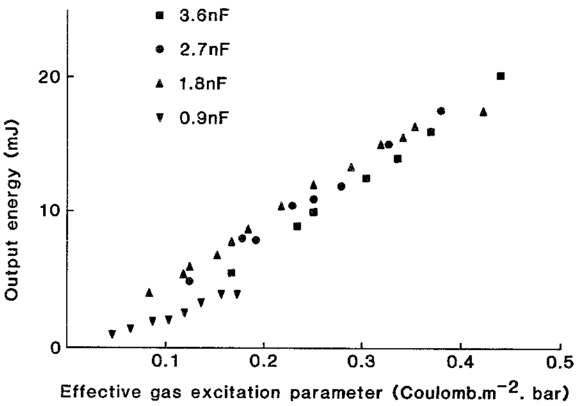

Fig. 3. Variation of output pulse energy with effective excitation parameter [$C \cdot m^{-2}$ bar] for 6.25% CO_2 mixture and a range of source capacitors (see text). (Charging voltage 40 kV)

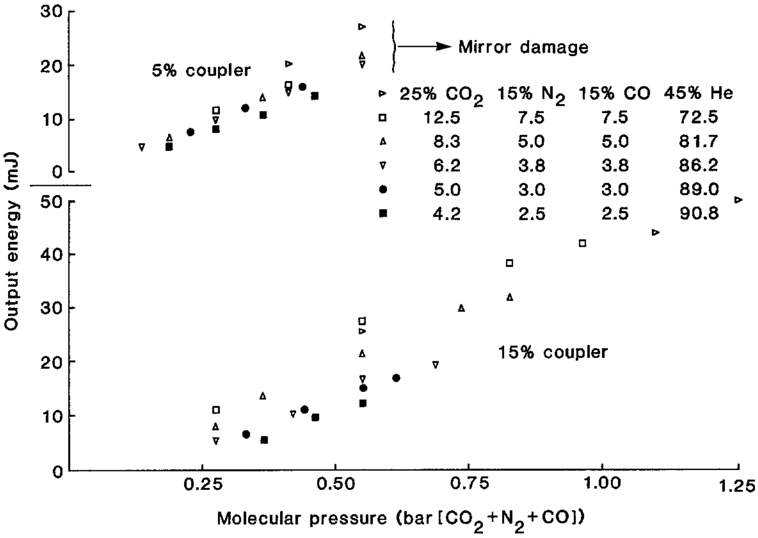

Fig. 4. Laser pulse energy versus partial pressure of molecular gas components $[CO_2+N_2+CO]$ for various He dilutions and different optical output coupling. The ratio CO_2:N_2:CO(5:3:3) the source capacitance (2.7 nF) are constant

for a given gas mixture. This emphasises that the total vibrational excitation is *source charge dependent*, not *source energy dependent*, and suggests that a suitable scaling parameter (at constant charging voltage) will be source capacitance per unit discharge cross-section multiplied by total gas pressure $[\mathrm{C.m^{-2}\cdot bar}]$. Figure 3 plots experimental results showing that this is indeed a reasonable scaling parameter for pressure of 1–6 bar and a range of source capacitors (0.9–3.6 nF), and much superior to the mode of analysis shown in Fig. 2b. Of course, the linearity displayed by Fig. 3 is not rigorous; there are secondary superelastic-collisional effects (see the end of this section). Nevertheless, $[\mathrm{C.m^{-2}\cdot bar}]$ is a useful indicator of expected laser output energy at various pressures, for a particular gas mixture.

Furthermore, for limited changes of gas mixture, a similar argument can be applied. Figure 4 illustrates the results of Fig. 1 (15% output coupling) as well as some results obtained with 5% coupling; the laser output energy is plotted against the total molecular gas pressure (CO_2+N_2+CO) where the ratio of CO_2:N_2:CO, and the source excitation capacitance, were kept constant, but the ratio of the total molecular content to helium content was varied. The total pressure was varied over the range 1 to 6 bar with molecular content ranging from 55% to 9.2%. Clearly the laser output energy varies roughly linearly with $[CO_2+N_2+CO]$. This is initially surprising, since the theory of McClellan et al. [6] predicts and E/n variation as $([CO_2+N_2+CO]p^{-1})^{2/3}$, implying a three-fold variation of E/n over the range of our measurements. The results therefore suggest that in the gas mixtures studied here, the excitation rates k_1, k_2, and k_3 vary only slightly with E/n, and that most important is the amount of the non-helium (molecular) gas fraction. This is reasonable since the dominant electron energy loss processes in such mixtures involve vibrational excitation and so both the energy absorbed in the discharge and the useful vibrational excitation are both closely dependent on the amount of the (vibrationally excitable) molecular component.

Figures 3 and 4 jointly indicate that the most useful laser output scaling parameter is $\mathrm{C\cdot m^{-2}}$ $\cdot\,[CO_2+N_2+CO]_{\mathrm{bar}}$.

Smith and Mellis [4] have recently pointed out that in the CO_2 TEA laser electron superelastic deexcitation of vibrational excitation must be considered and with even quite normal excitation conditions a typical atmospheric pressure laser is quite considerably excitation saturated, in that the upper laser level population is not significantly increased with further excitation, because both the electron excitation and deexcitation rates are increased. The degree of saturation is dependent on the vibrational excitation per particle, so the effective input parameter should be $(\mathrm{C\cdot m^{-2}}\cdot[CO_2+N_2+CO]_{\mathrm{bar}})\times[CO_2+N_2+CO]_{\mathrm{bar}}^{-1}\equiv\mathrm{C\cdot m^{-2}}$, and likewise the output energy should be normalised to unit pressure of the molecular component. This is illustrated in Fig. 5 for two sets of results (with the 15 and 5% couplers): the saturation effect can be clearly seen in both cases. Thus it is desirable to restrict the source capacitance per unit electrode area to $\lesssim 0.15\,\mathrm{C\cdot m^{-2}}$.

This very simple model seems to explain all the main features of the parametric behaviour of the laser output energy, and we have further determined that with other gas mixtures and for other charging voltages it is still a reasonable description. The main simplifications in the model are the assumed invariance of E/n with pressure and the need for the discharge impedance r' to be very much less than the source impedance R. We have already noted that the former is a reasonably good approximation over a wide range of circumstances,

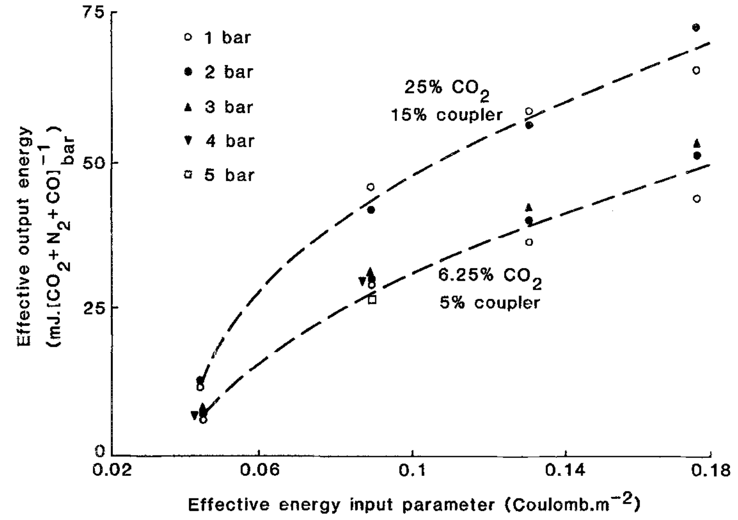

Fig. 5. Laser output energy (normalised to unit molecular gas pressure) against effective excitation parameter [$C \cdot m^{-2}$] for 25% and 6.25% CO_2 mixtures at various total pressures

while the latter is likely to become only very approximate as the pressure (and discharge impedance) increases further (as noted by Midorikawa et al. [1] in the 5–10 atm region). Although the impedance mismatch is very *convenient* for simple modelling it is of course *very bad* for the overall laser efficiency because it degrades the transfer of energy from the power supply to the gas discharge. It is best to limit the source capacitance per unit electrode area to minimise superelastic deexcitation, and maximise the total molecular partial pressure to maximise the gas impedance and hence total vibrational excitation within the limits set by discharge instability. In the extreme $r' \rightarrow R$ (with higher pressures and high molecular contents) and the vibrational excitation will no longer increase with pressure, but the laser energy may still slowly increase because then increasing the molecular content (by increasing the molecular fraction or the total pressure) will then reduce the superelastic saturation effect.

3. Discharge Instability Boundary

It is clear from Fig. 1 that at higher pressures the output energy which can be achieved is limited because the discharge becomes unstable and arcs unless the gas mixture is made helium rich. It was observed that higher charging voltages allowed stable operation to higher pressures (Fig. 6); i.e. for a given pressure the discharge instability boundary is a "minimum voltage" boundary. The arcing beyond this boundary came late in the discharge current pulse and can be associated with the current ringing (see [2] for further details of this mismatch-created-situation). It might be suspected that inadequate preionisation contributes to this instability, but seeding the gas mixtures with trace amounts of the low ionisation potential additive TPA did not significantly shift the arcing boundary, although the laser energy was increased (indeed the results of Fig. 6 are for a situation with some TPA additive and hence the energies are greater than those of Fig. 1). Thus the *variation* of this instability with applied voltage is probably due to variation of the discharge ringing characteristics with voltage and will probably vary from system to system and be eliminable in principle, or at least greatly reducible, with a shorter pulse with less ringing.

Figure 7 shows the effect of changing the value of the charging capacitor C_1 on the instability boundary. There appear to be two aspects to the behaviour: first, the instability boundary is not affected if the discharge voltage and C_1 values are kept low; the behaviour is like that of Fig. 6. But second, and quite distinctly, there is an apparently energy associated limitation: as the voltage is increased, so the maximum pressure of

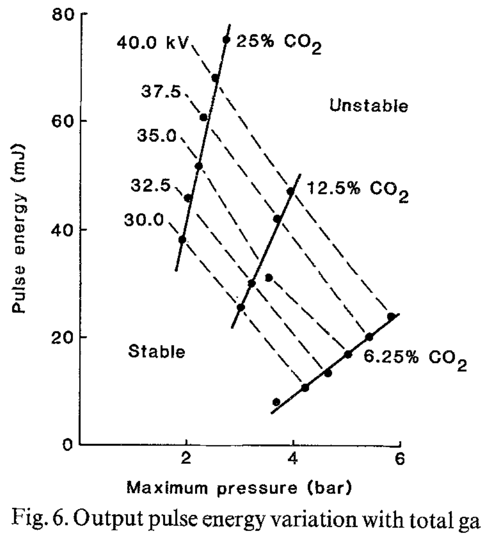

Fig. 6. Output pulse energy variation with total gas pressure for 3 mixtures showing discharge arcing boundaries for various charging voltages. Source capacitance 2.7 nF

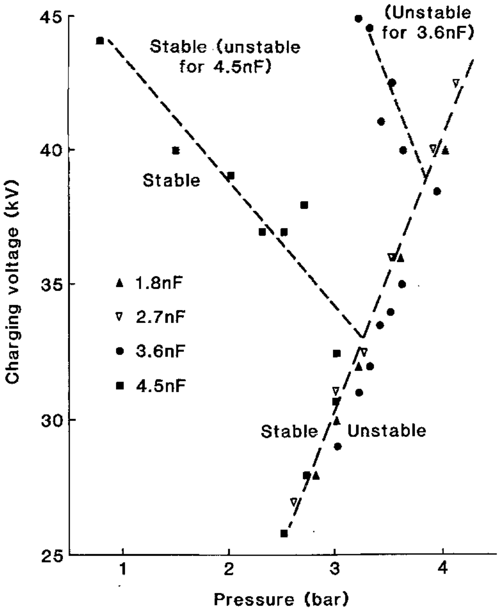

Fig. 7. Effect of changing source capacitance on discharge arcing boundaries in 12.5% CO_2 mixture. For the 1.8 and 2.7 nF capacitors no "maximum voltage" instabilities were observed up to the 45 kV power supply limit

operation starts to decrease rather than increase further. The onset of this change varies with the value of C_1, occurring at lower values of voltage with larger values of C_1. It is reasonable to hypothesise that this is an energy limitation associated with a thermal discharge instability. But the situation is complicated because changing the supply voltage affects both the energy loading and the exact pattern of delivery of the energy to the discharge (ringing, etc.).

Figure 8 further explores the hypothesis of a thermal discharge instability: it is a plot of the instability boundary in terms of the *molecular* (CO_2+CO+N_2) *partial pressure* and the *total* gas pressure. As we have seen the molecular concentration should be a good measure of the *actual* discharge energy loading, not just a measure of the energy available from the supply capacitor (Sect. 2). Results are plotted for the standard laser gas mixture, the mixture with added TPA and the mixture with extra N_2 content: in all cases for various helium dilutions. It is immediately apparent that all the points fall approximately on one boundary and (*a*) $[CO_2+N_2+CO]$ and thus probably the gas energy loading is the controlling parameter and (*b*) the instability boundary is not altered with enhanced preionisation and so is not preionisation controlled. The dashed line in Fig. 8 is not drawn as a best fit to the points, but is a 1/pressure line which seems to fit the variation of the instability boundary with pressure quite well.

To summarise: the arcing instability boundary appears to be dependent on the total heavy particle or molecular concentration, which can be associated with the *actual* gas energy loading. The maximum permissible molecular total concentration (not just fraction) decreases linearly with the total gas pressure. We are not aware of any predictions in the literature as to the pressure dependence of discharge thermal instability processes.

4. Optimum Laser Operation

It is clear from Fig. 4 that the laser output energy is mainly dependent on the total CO_2 concentration (or more precisely the total molecular concentration). But Fig. 8 shows that the molecular concentration achievable with arc-free operation increases as the pressure is decreased, with a limit as the total pressure approaches the molecular partial pressure (zero helium limit): this is indicated by the solid line in Fig. 8. Clearly the highest energies (and peak powers) should be obtained in the 1.5 to 2.5 bar region. Exact performance and optimum operating conditions will depend on the gas

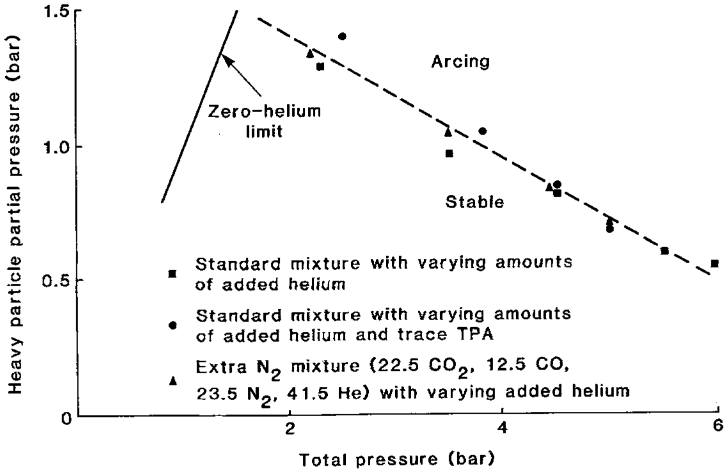

Fig. 8. Position of instability boundary in terms of molecular partial pressure $[CO_2+N_2+CO]$ and total gas pressure for 3 different mixtures at constant charging voltage (40 kV). The critical pressure for the onset of arcing depends only on molecular content, irrespective of the He dilution or mixture ratios

mixture, etc., and we have not attempted detailed parametric study. But we have obtained output energies >100 mJ and have determined that performance is improved by (*a*) adding a low ionisation potential additive such as TPA, (*b*) increasing the amount of nitrogen in the mixture (above that of the standard mixture), and (*c*) using $\approx 10\%$ hydrogen rather than helium as the atomic constituent.

If frequency tunability of the output is required such that operation at ~ 10 bar is required; then Fig. 8, etc., shows that the output performance will be a factor of ≈ 4 to 6 poorer than that achievable with the optimum 1.5 to 2.5 bar, or ≈ 3 to 5 poorer than that achievable at atmospheric pressure.

5. Photo Pre-Ionisation

It was tentatively concluded in Sect. 3 that the discharge instability boundary in the miniature TE laser is not directly due to inadequate pre-ionisation, both because it does not vary with the addition of a low ionisation potential dopant and because the instability develops late in the main discharge current pulse. However, adequate preionisation must always be an essential requirement and in this section we examine how the pre-ionisation may vary with pressure, firstly in terms of the pre-ionisation produced and secondly in terms of the pre-ionisation required for satisfactory glow discharge initiation.

Consider first the pressure dependence of the photo pre-ionisation electron density produced in the gas volume and its dependence on the three factors: the uv photon production at the source spark, the photon transmission through the gas, and the photo-ionisation electron production in the laser discharge volume. It is convenient to treat these in reverse order: –

(i) It had recently been established [7, 8] that the photo-ionisation in the laser gases is due to minority species (initial gas impurities such as propene and species created in the discharged gases such as NO and NO_2). The concentration of these species should scale with pressure, i.e. $\propto p$.

(ii) Transmission of the uv photons responsible for the photo-ionisation is strongly inhibited by CO_2 absorption, and photo-ionisation is a single step process due to photons at ~ 120 nm in a relative transmission window with an absorption coefficient of $\sim 2\,\mathrm{cm}^{-1} \cdot \mathrm{atm}\,CO_2^{-1}$. Thus increasing the pressure (and CO_2 partial pressure) will decrease the transmission (dependent on the % CO_2, distance of travel etc.) typically by a factor of 3 to 5 as the pressure is increased from 1 to 5 bar in a miniature TE laser (but by several orders of magnitude in larger devices).

(iii) Babcock et al. [9] have suggested that the actual ~ 120 nm emission from the source arc is $p^{1/2}$. We have not been able to directly examine this but we have made a series of indirect observations. First, using a fast photomultiplier tube and fibre-optic (sensitive in the visible region, not uv) we have checked the variation of spark emission intensity with pressure (with constant electrical discharge source) both in the high pressure laser over the range 1 to 5.3 bar and in the photo-ionisation chamber system (with the co-operation and assistance of S.J. Scott) from 0.25 to 1 bar. The emission increases very slightly with pressure (less than $p^{1/2}$). Second in the photo-ionisation chamber S.J. Scott has established (assuming the results of (i) and (ii) that for typical high pressure lasers the emission will increase moderately with pressure ($p^{1/2}$ or less). We conclude that for modelling purposes it is reasonable to assume that the emission increases a little with pressure, but probably not as much as $p^{1/2}$.

Overall, from (i), (ii), and (iii) we conclude that the photo-electron yield in the miniature TE laser volume will vary little with pressure, if anything increasing slightly.

It is, of course, interesting to look at the theoretical literature to ascertain the expected minimum pre-ionisation electron density $n_e(o)$ required as the pressure is increased. Unfortunately the main works (Palmer [10], Karnyushin et al. [11, 12], Levatter and Lin [13], Hertziger et al. [14]) are far from comprehensive and, of the limited subject area they treat, they are far from being in mutual agreement. All the authors (following Palmer) regard as crucial the need for the heads of the initial streamers (radius r), to overlap before the local electric field becomes too (critically) distorted. The critical size of the head r_c then depends on whether it is thought that the growth of r is free diffusion controlled or is directly field dependent, and if the time to critically is pressure (p) dependent. The analyses are far from reassuring, but suggest $r_c \sim p^{-1}$ or $r_c \sim p^{-1/2}$. (The much quoted analysis of Levatter and Lin which leads to the latter seems to assume that the avalanche path length and the free diffusion velocity are independent of the field). There is then agreement that $n_e(o)$ should be sufficient that at criticality the heads of adjoining streamers overlap, so $n_e(o) > r_c^{-3}$ thus $n_e(o) \sim p^3$ (or $p^{3/2}$).

There is also the (equally important) need for the initially established electron volume rapidly advancing towards the anode to be controlled [11, 15] such that there is sufficient (photon, etc.) feedback to the gas as far back as the cathode for complete Raether-Meek streamers to develop within the development time of V_d/d where V_d is the drift velocity and d the gap width. The pressure dependence of this has not been established. So the overall theoretical situation is very uncertain: but $n_e(o)$ probably increases very rapidly with pressure.

We conclude this section then on a rather unsatisfactory note. Theoretically the preionisation density required for uniform glow discharge initiation increases very rapidly with pressure, whereas the preionisation density produced in a laser increases only very slightly with pressure. But the weight of the rather circumstantial evidence is that such a lack of preionisation is not the chief contributor to the actual instability boundary (at least in the miniature TE laser with a very strongly preionised system). Instead it is necessary to look at the pressure dependence of the rate of development of any instability in an initially well developed glow discharge.

6. Sealed Laser Operation

We have previously fully established that in atmospheric pressure TE lasers the major limitation on long life sealed gas operation is the build-up of oxygen from CO_2 dissociation and the subsequent attachment of electrons

$$e + O_2 + M \rightarrow O_2^- + M$$

followed by charge exchange reactions. This attachment process affects both the preionisation (reducing the preionisation electron density in the delay period between the preionisation and main discharge pulses [16]) and the main discharge (altering the charge particle balance and directly leading to instability [17]). The concentration of oxygen is best controlled by either using a heterogeneous catalyst (such as hot platinum) to reform CO_2 or a homogeneous catalyst (such as CO).

We have examined the pressure dependence of the amount of oxygen which can be tolerated in typical laser mixtures in the high pressure laser. Figure 9 shows the results obtained. As the pressure is increased the tolerance for oxygen very rapidly decreases, from up to 15% at 1 bar to 0.1% or less at 5 bar. The exact amount depends on the gas mixture, CO_2 rich mixtures having lower tolerances. Also shown are some re-plotted sub-atmospheric pressure results of Aleinikov et al. [18] showing similar behaviour.

Of course, not only will the results vary with mixture, but also with the nature of the discharge circuit, adequacy of preionisation etc.; and overall we observe a pressure dependence of $\approx p^3$ to $\approx p^4$. The three body attachment reaction will have a p^2 dependence, and so can contribute significantly to the change of oxygen tolerance with pressure. There is clearly also at least one other significant factor, possibly the pressure dependence of the rate of development of the discharge instability. The suggestion is that the discharge instability develops more rapidly at higher pressures (this would also be consistent with the results of Sect. 3) and so become less tolerant of any fluctuations such as oxygen-generated negative ion perturbations.

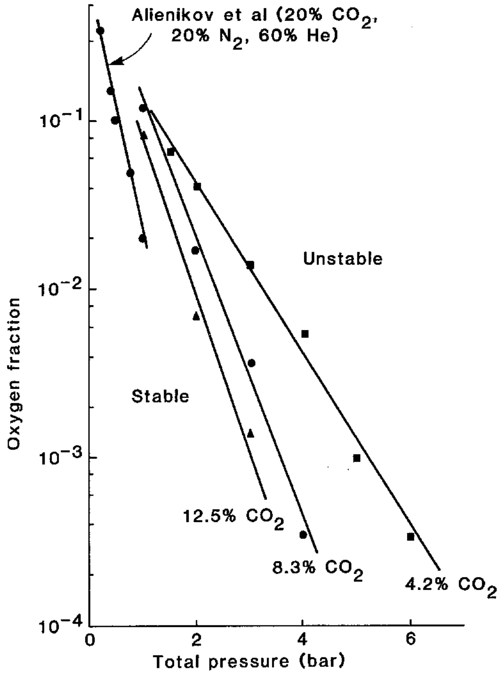

Fig. 9. Stable discharge tolerance to added oxygen as a function of total gas pressure for CO_2:N_2:CO:He mixtures in ratios 12.5:7.5:7.5:72.5, 8.3:5.0:5.0:81.7, and 4.2:2.5:2.5:90.8. Also plotted are the results of [18] for a 20:20:60 mixture

To make a sealed long-life single gas fill high-pressure laser thus requires the maintenance of a very low oxygen concentration. There are two ways to proceed:

(i) Use heterogeneous catalysis, probably with a slow gas re-cycle loop such as that recently described by Smith et al. [3]. This should lead to little extra complexity or volume for large systems and should be entirely effective.

(ii) Use homogeneous catalysis (such as CO or CO + H_2). This is satisfactory for atmospheric pressure devices, but even at atmospheric pressure the highest peak power mixture systems require additional catalysis [3]. To our knowledge no long-life supra-atmospheric pressure lasers have been successfully made and we have now investigated to what pressures the homogeneous catalysis technique can be successfully utilised for miniature TE devices.

We have run a series of 12 sealed lifetime tests in the high pressure laser. The standard 25% CO_2, 15% CO, 15% N_2, 45% He mixture produces long-life operation ($>10^5$ pulses) at 1 bar, but at 2 bar main discharge arcing commences after 1.0 to 1.5×10^3 pulses when the CO_2 dissociation (measured with a MS10S mass spectrometer) has reached $4 \pm 1\%$. To obtain longer life operation it is necessary to lower the percentage of CO_2 and increase the CO/CO_2 ratio to produce a decreased $[0_2]$ production. For instance, at 5 bar long-life operation ($>10^5$ pulses) was obtained with 3% CO_2, 6% CO, 2% N_2, 89% He. It was also found that the addition of a small amount of hydrogen further restricted the dissociation of CO_2 and a mixture with rather more CO_2 could be successfully operated: thus in 5 bar of 5% CO_2, 5% CO, 3% N_2, 2% H_2, 85% He the dissociation was $<5\%$ after 10^5 pulses. With such a mixture operating at 1–2 pps and the 5% output coupler the output energy was ~ 12.5 mJ initially, decreasing to ~ 6 mJ after 10^5 pulses.

To summarise: long-life sealed operation using homogeneous catalysis has been successfully demonstrated at up to 5 bar total pressure, most successfully with a combination of CO and H_2 as catalyst. All the arcing boundary limitations observed with different mixtures and pressures were quantitatively consistent with the arcing threshold being due to the creation of CO_2 dissociation produced oxygen, as described in Fig. 9.

7. Summary and Conclusions

1) As the pressure is increased from atmospheric to ~ 5 bar the laser pulse shape is little altered but both the peak power and total energy increase significantly with the pressure. Quantitatively the *output energy* scales with the product of the source charge input per unit electrode area $[C \cdot m^{-2}]$ and the total molecular partial pressure $[CO_2 + N_2 + CO]$ in bar. The main features of this behaviour are explicable in terms of the variation of the actual energy input into the gas with pressure (although the source excitation remains constant) due to the changing impedance of the discharge (largely dependent on the vibrational excitation of the molecular gas fraction).

2) It is essential to have adequate uv photo preionisation. We find that the photo-electron yield in the miniature TE laser gas varies little with pressure, if anything increasing slightly (but in a larger dimensioned laser there may be a considerable decrease). Theoretically the required pre-ionisation *to establish* a uniform glow discharge increases rapidly with pressure, but our experimental results tend to suggest that our sliding-spark pre-ionisation is adequate and the observed discharge instabilities are due to subsequent collapse of the glow discharge, not its inadequate initiation.

3) With a fresh gas fill (no dissociation products) the gas becomes more unstable as the pressure is increased, such that arc-free discharges can only be obtained in helium-rich mixtures. The maximum permissable molecular *total concentration* (not just fraction) decreases linearly with total gas pressure. The discharge arcing appears to be associated with a more rapid development of instability in the main glow discharge at higher pressure.

4) As a consequence of 1) (the increase of output energy with $[CO_2 + N_2 + CO]$ pressure) and 2) (the improvement in discharge stability with decrease in pressure) there is an optimum operational region of 1.5 to 2.5 bar. If operation at ~ 10 bar is required (for continuously tunable output) then we would expect the output to be $\approx \times 3$ to $\times 5$ poorer than that achievable at atmospheric pressure and $\approx \times 4$ to $\times 6$ poorer than that achievable at the optimum 1.5 to 2.5 bar.

5) With sealed gas operation the production and presence of oxygen becomes of overriding importance. The discharge instability boundary has a $\approx p^3$ to p^4 pressure dependence (depending on the gas mixtures etc.). This appears to be due to oxygen-attachment accentuation of the main discharge instability (3 above). Long life sealed operation ($>10^5$ pulses) has been obtained using homogeneous catalysis (CO and H_2 added) at up to 5 bar, but only in helium-rich mixtures, resulting in low levels of laser output. Alternatively, heterogeneous catalysis can be used to obtain in sealed systems performance very similar to that achievable in the fresh-gas systems.

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