FEBRUARY 1978 LRP 135/78

THE INFLUENCE OF ADDITIVES AND CONTAMINANTS IN TEA CO_2 LASER DISCHARGES EVALUATED BY ELECTRICAL MEASUREMENTS

A. Lietti

FEBRUARY 1978 LRP 135/78

THE INFLUENCE OF ADDITIVES AND CONTAMINANTS IN TEA CO_2 LASER DISCHARGES EVALUATED BY ELECTRICAL MEASUREMENTS

A. Lietti

Abstract

Observation of the electrical characteristic of the glow discharge allows easy study of the influence of additives and contaminants. Moreover, when arcing occurs, interesting information is obtained by observing the damping of the arc current. The efficacy of low-ionization organic additive to prevent arcing is confirmed, and the deleterious influence of oxygen contamination is demonstrated. This result accounts for the technical rule of preventing air entering the discharge vessel. Increased output peak power is observed, resulting from the use of suitable additive.

The use of low-ionization-potential organic additives is an established means of preventing arc discharges in TEA CO_2 laser operation[1,2]. The influence of these additives has been recently evaluated by observing the photoionization spectra[3] and the electron density[4]. Little information, however, can be found about the role of contaminants. Denes and Lowke[5] considered the influence of water vapor.

This Letter reports evaluation of additives and contaminants by simple electrical measurements.

The measurements were performed on a modified version of a double discharge laser, already described elsewhere[6]. Following Richardson et al.[7], the preionization is produced by a two dimensional array of high voltage arcs. The dielectric plate, which supports the array, is located behind a flat grid electrode. The second electrode has a Chang[8] profile. The interelectrode distance is 5 cm, and the surface covered by the regular discharge is 600 cm^2. The discharge starts 2μsec after the beginning of the preionization. A Marx generator produces the high voltage pulse, which ranges from 50 to 80 kV with a rise time of 150 nsec. The gas was flowing at the rate of 10 liter/min. First we observed the discharge at full power (80 kV pulse). Typical records of the current both in stable and unstable discharges are shown in Fig. 1. The pulse generator and the discharge gap form an electrical LCR circuit, where L = 0.4μH, C + 0.05μF, $R = R_i + R_g$. $R_i = 0.05\Omega$ accounts for the internal losses of the generator, R_g is the gap resistance. Note that, when normal glow discharge occurs, the circuit is more than critically damped. On the contrary, when arcing occurs, the decrease of R_g allows the circuit to oscillate.

When the discharge was stable, we measured the ratio of electric field to neutral gas density, E/N, as a function of the current density.

This way, for various gas mixtures, the quasistable operating value E_g/N was estimated[5]. In high pressure glow discharges the field is nearly constant for a typical range of currents. In this range, the minimum of the field derivative with respect to current defines E_g. Following Dyer and James[9] we observed R_g and ρ (the gas resistivity) at peak current as a function of peak current density i. In this experiment we noted that, when $i \approx 10$ A/cm^2, $\rho i \approx E_g$. When the voltage is raised to 80 kV, the laser works at full power, and the current density increases to the highest value, i_{max}. In this condition we measured the output energy of the laser beam by an absolute calorimeter[10].

When arcing occurs, as Fig. 1b shows, the current can be described with good approximation by :

$$i = i_o \exp\{-(\alpha+j\omega)t\}$$

That means that the arc behaves like a resistance. The gap resistance is now very low and can be deduced from the damping of the current.

$$R_g = 2\alpha L - R_i$$

Table I shows measured values of the discharge parameters for mixtures at atmospheric pressure with a constant CO_2/N_2 ratio. With the increase of the $(CO_2+N_2)/He$ ratio, both field and resistivity increase, and current decreases, in agreement with previous measurements[8,9].

A simple model of the high pressure glow discharge, which considers the ionization attachment rate equations, explains the experimental results and shows that the decrease of current is connected with a decrease of electron density. The experiment shows, however, that beyond a certain limit, instability occurs. The influence of the addition of electronegative gases like oxygen and of low-potential ionizations chemicals can be explained by the variation of the electron density, n_e.

In this experiment addition of 1% of oxygen always produces instability. We investigated the following chemicals : tri-n-propylamine, p-xylene and di-n-butylamine (ionisation potentials 7.23; 8.44; 7.69eV). The first one is by far the most effective, followed by the second one, even if the quantity of tri-n-propylamine which was introduced in the mixture was ten times less than the quantity of the other additives. The difference cannot be explained by considering the ionisation-potential hierarchy alone. Spark-source emission spectra[3], as well as complex molecular energy bands of all the components of the mixture have to be taken into account.

Turning now again to the information that can be deduced from Table I, we note that in the glow discharges, the parameters most sensitive to seedant addition are i_{max} and R_g. Both are measured when the pulse generator is charged to the highest voltage.

Considering the electrical circuit, this situation approaches the case of constant voltage generator. The parameters E_g/N and ρ_o, on the contrary, are consistent with the reverse situation of a constant current generator. They are chiefly functions of the gas composition and show little dependence on experimental conditions, but they are less sensitive to small variation of the gas mixture than the parameters i_{max} and R_g.

These parameters, however, show some dependence on experimental conditions, and are chiefly useful as differential indicators. The behaviour of the gap resistance R_g, both in glow and in arc discharges deserves some comments. Fig. 2 shows the variation of R_g during the transition from glow to arc and from arc to glow, which results from the addition of destabilizing and stabilizing substances. During the glow phases, the variations of R_g are consistent with opposite variations of i and n_e, according to the model discussed above. The opposite behaviour of

R_g in arc discharges is the consequence of a different physical situation. Arc discharges were investigated in the past[11,12], and taking into account these previous studies, a preliminary calculation explains the behaviour of the gap resistance by the variation of arc temperature. Addition of low ionization potential chemicals lowers the arc temperature and increases R_g. The opposite effect occurs with the addition of electronegative gases. Note that the numerical result depends somewhat on the boundary conditions around the arc column. This imples a partial dependence on the gas preionization. We conclude that the gap resistance is a good differential indicator in studies concerning the influence of additives and contaminants, both in stable and unstable discharges.

Consider now the destabilizing influence of oxygen. It explains the experimental observation that laser discharges are often unstable when the apparatus had been exposed to air, or when the vessel is leaky. In this case, a continuous flow of gas is required, even when the apparatus is not working. After exposure to air, we observed arcing. Measurements showed a continuous increase of R_g with time, until stable discharge occurs, about two hours after the exposure. Wall and electrode absorption could explain this long term contamination. Water vapor was also shown to have a deionizing effect[5]. We noted, however, that contamination occurs even with dry air.

A few words about the influence of organic additives on laser output. Table I shows that the additives improve the laser efficacy because they allow an increase of CO_2 content in the mixture. We did not observe any increase of output when additive was added to a given gas mixture, but we noted an improvement of the beam homogeneity. We did not try, however, to maximize the output by increasing the field.

The author wishes to thank H. Van den Bergh, A. Heym and J.-M. Mayor for useful discussions. He is indebted to the laser group of the CRPP for general assistance with the experiment, and to M. Green for reading the manuscript.

This work was supported by the Swiss National Science Foundation.

References

1 R.L. Schriever, Appl. Phys. Lett. 20, 354 (1972)

2 Laurence E. Kline, L.J. Dener, and M.J. Pechersky, Appl.Phys.Lett. 29, 574 (1976)

3 H.J.J. Seguin, D. McKen, and J. Tulip, Appl.Phys.Lett. 29, 110 (1976)

4 E. Morikawa, J.Appl.Phys. 48, 1229 (1977)

5 L.J. Denes and J.J. Lowke, Appl.Phys.Lett. 23. 130 (1973)

6 P. Boulanger, A. Heym, J-M. Mayor, and Z.A. Pietrzyk, J.Appl.Math & Phys. 24, 439 (1973)

7 M.C. Richardson, K. Leopold and A.J. Alcock, IEEE J. Quantum Electron, QE-9, 934 (1973)

8 T.Y. Chang, Rev.Sci.Instrum. 44, 405 (1973)

9 P.E. Dyer and D.J. James, J.Appl.Phys. 46, 1679 (1975)

10 P. Boulanger, A. Heym, J-M. Mayor and Z.A. Pietrzyk, J.Phys. E 6, 559 (1973)

11 H. Maecker, Z. Physik 136, 119 (1953)

12 B. Busz and W. Finkelburg, Z. Physik 138, 212 (1954)

Figure Captions

Fig. 1 Typical laser current pulse shapes

a) Regular glow discharge (I = 5kA/div, t = 0.2μsec/div).

b) Arc discharge (I = 15 kA/div, t = 0.5μsec/div).

Fig. 2 The gap resistance R_g during the transitions

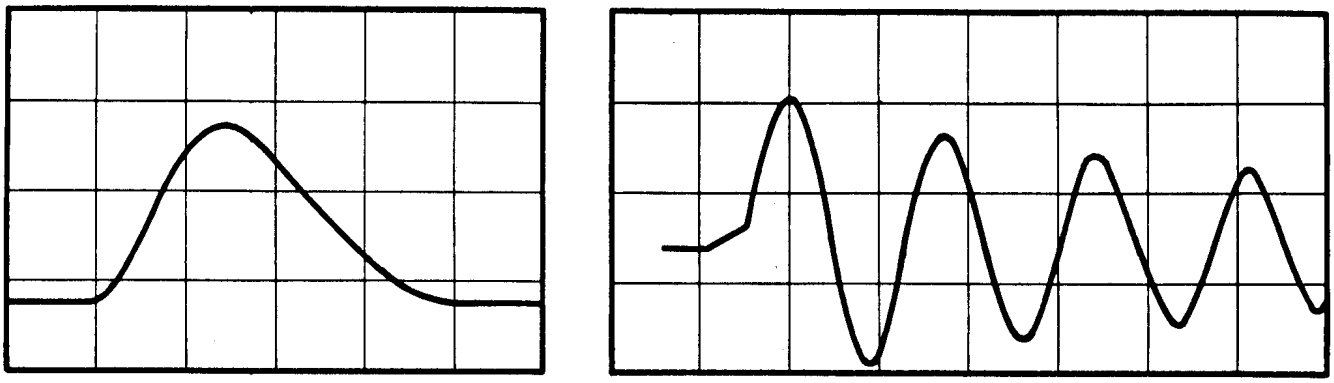
a) Transition from glow to arc in the CO_2/N_2He = 10/10/80 mixture by addition of oxygen.

b) Transition from arc to glow in the $CO_2/N_2/He$ = 30/30/40 mixture by addition of tri-n-propylamine.

TABLE I

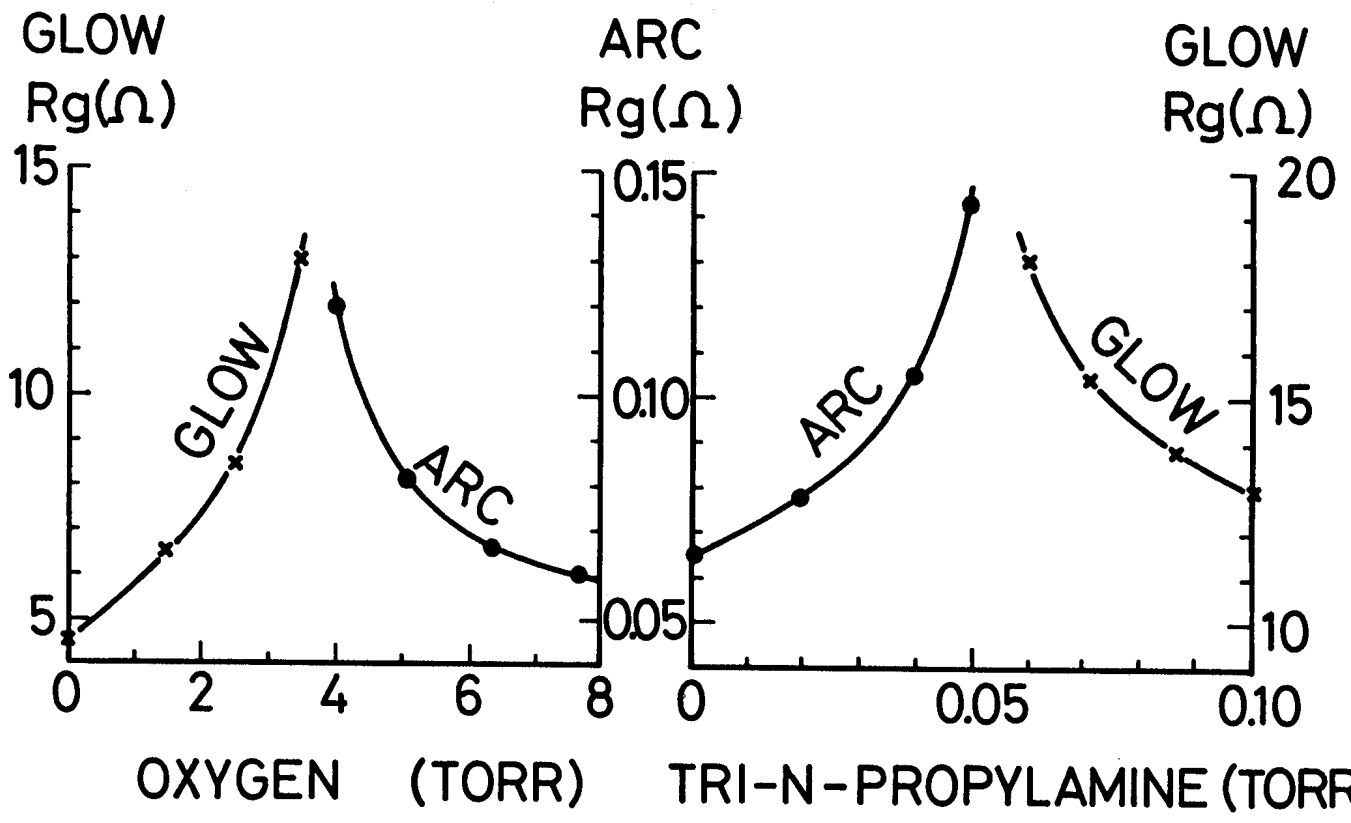
Measured values of the discharge parameters. E_g/N is the normalized quasisteady field. ρ_o is the gas resistivity at the peak current density of 10 A/cm^2. i_{max}, W and R_g are the peak current density, the output beam energy and the gap resistance resulting from the input pulse of 30 kV. A_1 means addition of tri-n-propylamine at the partial pressure of 0.1 Torr, A_2 of p-xylene at 1 Torr, A_3 of di-n-butylamine at 1 Torr, O_2 or oxygen at 10 Torr.

Gas Composition CO_2 / N_2 / He	E_g/N (10^{-16}V cm^2)	ρ_o (Ω cm)	i_{max} (A/cm^2)	W (J)	R_g (Ω)
10/10/80	3.59	970	21	12	4.5
10/10/80 + A_1	3.30	890	29	12	2.5
10/10/80 + O_2		arc			0.060
15/15/70	4.17	1130	14	14	8.0
20/20/60	4.88	1320	11	15	10.5
20/20/60 + A_1	4.44	1200	17	15	7.5
20/20/60 + A_2	4.63	1250	13	15	9
20/20/60 + A_3	4.79	1290	12	15	10
20/20/60 + O_2		arc			0.060
20/20/60 + O_2 + A_1		arc			0.060
25/25/50		arc			0.075
25/25/50 + A_1	5.13	1380	13	16	10
25/25/50 + A_2	5.63	1520	10	16	11
25/25/50 + A_3		arc			0.080
30/30/40		arc			0.065
30/30/40 + A_1	5.90	1600	10	17	13
30/30/40 + A_2		arc			0.070



(a) (b)

FIG 1



(a) (b)

FIG 2