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Effective swarm parameters and transport coefficients in CO₂ laser mixtures

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Abstract. A pulsed electron swarm technique has been used to obtain effective attachment and ionisation cross-sections as well as electron drift velocities in mixtures of CO₂ laser interest. In binary CO₂:N₂ mixtures, below a reduced electric field of $E/N=60$ Td ($1 \text{ Td}=10^{-17} \text{ V cm}^2$), attachment was the principal ion production process. (N is the total gas number density.) The reduced attachment coefficients η/N measured were small ($< 10^{-19} \text{ cm}^2$) and in excellent agreement with numerical predictions.

In ternary mixtures of He:CO₂:N₂, positive ion formation described by Townsend's first ionisation coefficient α played a more important role over the same range of E/N . The reduced coefficient for total ion formation, $(\alpha+\eta)/N$, was measured in 12:4:1 and 8:1:1 mixtures and found to be in good agreement with numerical calculations.

Electron drift velocities in both the binary and ternary mixtures were determined using a time-of-flight technique. Overall agreement with previous experimental and numerical results was good.

1. Introduction

Optimal operation of CO₂ lasers requires detailed knowledge of the transport parameters which control the development of a gas discharge. Numerous studies have been carried out to determine swarm parameters in *pure* gases, and several data compendia are available (e.g. Brown 1966, Dutton 1975). Modern CO₂ lasers, however, use *mixtures* of He, N₂ and CO₂, and surprisingly little experimental swarm data exist for the combinations of interest. A number of investigators (Sakai *et al* 1979, Lowke *et al* 1973, Nighan and Bennet 1969) have used numerical methods to predict swarm parameters in laser mixtures and to estimate the populations of the laser levels. Experimental determination of some of these calculated swarm parameters provide a valuable assessment of the accuracy of the numerical methods.

The work presented in this paper represents an experimental determination of the ionisation coefficient α/N , attachment coefficient η/N , and electron drift velocities W in mixtures of CO₂ laser interest over a wide range of E/N and total gas pressure. The method is similar to that previously reported by Nygaard *et al* (1979, 1978a, b) for attaching gases. The study of CO₂ mixtures presents additional difficulties since both ionisation and attachment are expected to be important over the E/N range of interest,

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and because of the small values of the coefficients. Details of the experimental procedures and the difficulties are given in the next section. The cases investigated were binary $\text{CO}_2:\text{N}_2$ and ternary $\text{He}:\text{CO}_2:\text{N}_2$ mixtures. In addition, some pure CO_2 and N_2 data were taken to check the experimental procedure. Resulting coefficients for attachment and ionisation as well as drift velocity of electrons are given in §3.

2. Experimental method

The method used in this work is the same as previously described in Brooks *et al* (1979). Figure 1 shows a block diagram of the experiment. The drift region is enclosed in an UHV chamber; typical background pressures are about 10^{-8} Torr after 24 h at 150°C . The gas mixtures to be studied are prepared in the chamber from high purity grade gases used as received. Light from a pulsed UV laser (Xe ion at 2315 \AA , 100 ns wide, or ArF at 1936 \AA , 17 ns wide) enters the chamber through a sapphire port, passes through a screened hole in the anode and strikes the cathode. A short burst of electrons is produced, which then drifts toward the anode under the action of the applied electric field. The voltage drop developed across the anode resistor R_a is sent to a unity gain high impedance buffer and recorded on the AEL model 9300 pulse and transient recorder. An on-line Smoke Signal Broadcasting (SSB) microcomputer is used for signal averaging and data analysis.

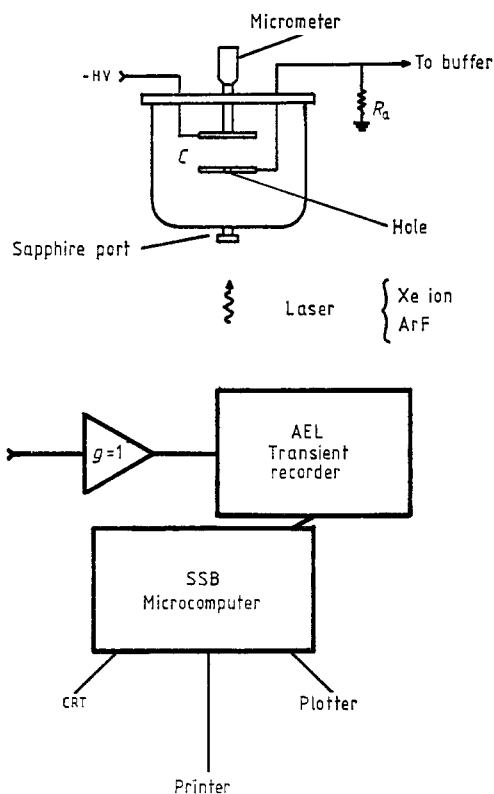


Figure 1. Schematic diagram of apparatus.

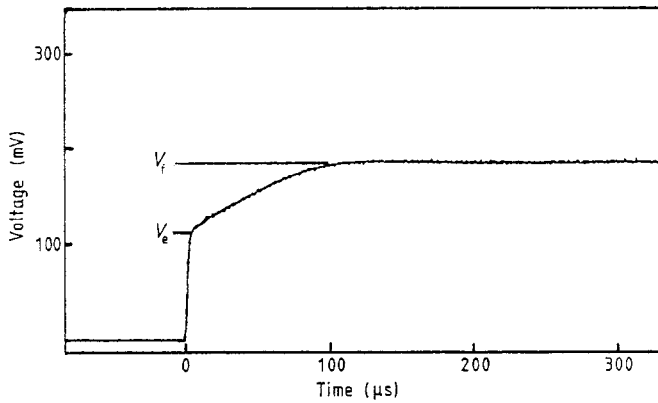


Figure 2. Typical integrated anode voltage transient obtained with $R_a = 10 \text{ G}\Omega$. The sharp initial rise is due to electrons and reaches an amplitude V_e at the electron transit time T_e . The subsequent slow rise is due to collection of negative and positive ions.

When the anode resistor R_a is very large, ($10 \text{ G}\Omega$), it has been shown by Brooks *et al* (1979) that the voltage transient consists of a fast electron component and a slower ion component, as illustrated in figure 2. In cases where electron diffusion and positive ion formation may be ignored, the *ratio* of the final voltage, V_f , to the voltage observed at the end of the electron transient, V_e is

$$R = \frac{V_f}{V_e} = \frac{\eta d}{1 - \exp(-\eta d)} \tag{1}$$

where η is the attachment probability per unit length, and d is the inter-electrode separation. The extension to the case when both positive and negative ions are formed is easily derived by a similar analysis and is given by

$$R = \eta d - \frac{(\alpha - \eta)d \exp [(\alpha - \eta)d]}{1 - \exp [(\alpha - \eta)d]} \tag{2}$$

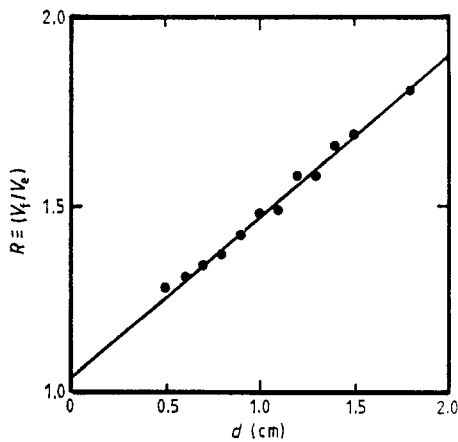


Figure 3. The ratio $R \equiv V_f/V_e$ plotted as a function of electrode separation in a He: CO₂: N₂ = 12:4:1 mixture at 41.6 Td.

It would appear from equation (2) that if the ratio R is measured at several values of d , the parameters η and $(\alpha - \eta)$, (and hence α) could be obtained by data fitting. In reality this proves to be impossible. Equation (2) may be expanded to second order to give

$$R = 1 + \frac{1}{2} (\alpha + \eta)d + \frac{1}{12} (\alpha - \eta)^2 d^2 + \dots \tag{3}$$

It follows from equation (3) that for $(\alpha - \eta)$ small, R is a *linear* function of the inter-electrode spacing d with slope $\frac{1}{2} (\alpha + \eta)$. The quantity $(\alpha - \eta)$ may be small either because $\alpha \approx \eta$ or α and η are both small. Figure 3 demonstrates typical data with R plotted against inter-electrode distance d . It is clear from this figure that R is a linear function of d . From the slope of curves such as this one, attachment and ionisation coefficients have been obtained.

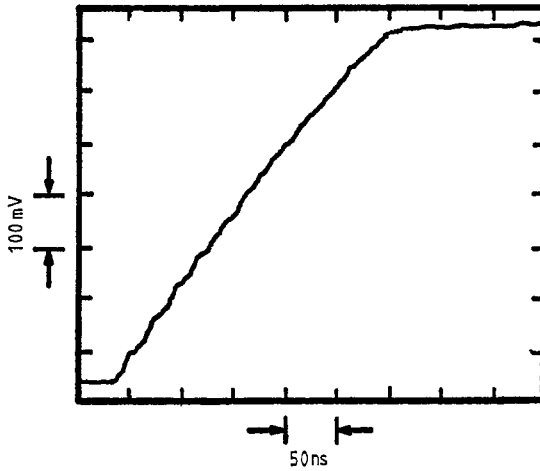


Figure 4. Integrated electron transient in a $\text{CO}_2:\text{N}_2=4:1$ mixture at 56 Td. In this example, the electron transit time is 316 ns.

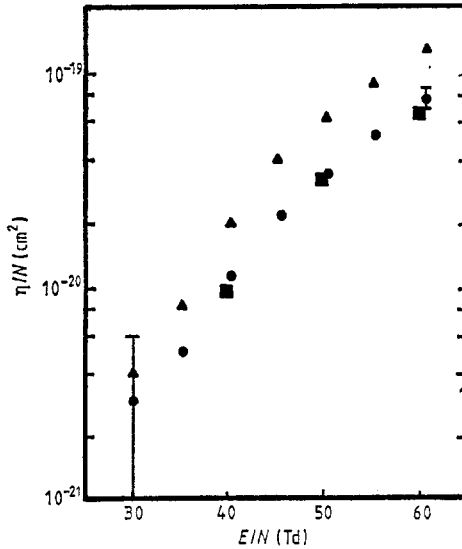


Figure 5. Attachment coefficient η/N as a function of E/N in pure CO_2 (●) and in a $\text{CO}_2:\text{N}_2=4:1$ mixture (▲). The filled squares represent theoretical calculations by Leland (1979).

One other piece of information which may be obtained in these experiments is the drift velocity of the electrons. In all drift velocity measurements the fast ArF laser was used, while either laser could be used in the attachment and ionisation experiments. It was shown above that when the anode resistor R_a is very large, a fast electron transient and a slow ion transient are observed. By greatly expanding transients such as that of figure 2 it is possible to resolve the electron transient temporally and to assign a transit time to the electrons. An alternative method (Sierra *et al* 1979) is to use $R_a = 50 \Omega$ and measure the electron current directly. Electron drift velocities determined by the two methods give identical results.

3. Results

3.1. CO₂:N₂ mixtures

In the present work a number of binary mixtures of CO₂:N₂ were studied. The mixtures ranged from pure N₂ to pure CO₂ in the pressure range 50–600 Torr. These binary

Table 1. Reduced attachment coefficient η/N , in CO₂ and (CO₂+N₂) gas mixtures. Results are accurate to $\pm 10\%$ unless indicated. E/N values are accurate to $\pm 3\%$.

E/N (Td)	η/N (10^{-21} cm ²)		
	100% CO ₂	80% CO ₂ +20% N ₂	20% CO ₂ +80% N ₂
30.3	3.9 ± 3	3.0 ± 3	$2.1 \pm \frac{2}{3}$
35.4	8.2	5.1	—
40.6	20	14	4.3
45.5	40	22	4.8
50.6	62	35	3.5
55.6	89	53	13
60.8	130	77	15

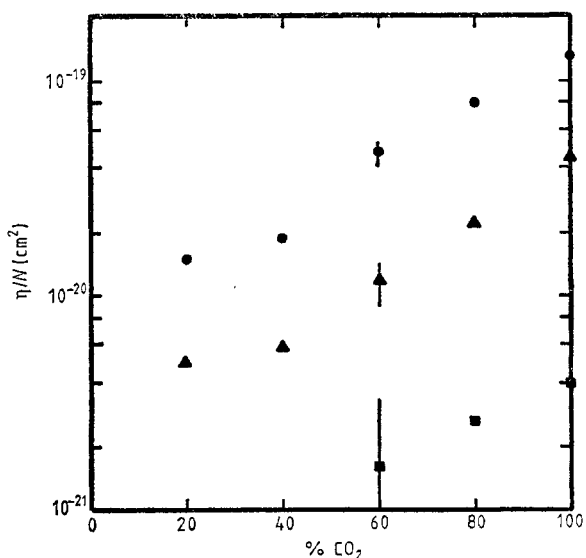


Figure 6. Effect of varying the mixture composition on the attachment coefficient at three different values of E/N . ●, 60.7 Td; ▲, 45.5 Td; ■, 30.3 Td.

mixtures are of importance for their use in CO₂ laser amplifiers (Leland *et al* 1978).

As discussed in the previous section, the observable quantity in the present experiment is the sum ($\alpha + \eta$). Over the E/N range 30–80 Td, attachment is expected to dominate over ionisation. Evidence for this assumption is given in figure 4. There, the electron transient for a 4:1 mixture at 56 Td is shown, for which $\eta > \alpha$ implies a downwards concave curvature such as that observed. For these mixtures data were only taken where $\eta \gg \alpha$, thus allowing the determination of η/N .

Figure 5 shows the attachment coefficient η/N plotted against E/N for pure CO₂ and for a mixture of 80% CO₂ + 20% N₂. Also shown are the results of a Boltzmann code for the latter mixture (Leland 1979). Excellent agreement is observed between the measured and calculated values. Table 1 gives the η/N data for the three mixtures studied; limits of error were assigned from the scatter in the data. Below 30 Td no ion transient was observed in any of the mixtures. The limit of detectability in the present experiments sets η/N at less than 5×10^{-22} cm² for E/N below 30 Td.

The effect of varying the mixture composition at constant E/N is shown in figure 6. There, the attachment coefficient η/N is plotted against mixture composition, where the

Table 2. Drift velocity of electrons in CO₂:N₂ mixtures. E/N values are accurate to $\pm 3\%$. Results are accurate to $\pm 5\%$.

E/N (Td)	$W(10^6 \text{ cm s}^{-1})$			
	CO ₂	20% N ₂ +80% CO ₂	80% N ₂ +20% CO ₂	N ₂
1.5	—	—	0.89	—
3.0	0.54	0.67	1.85	—
4.5	—	—	2.20	—
6.1	1.07	1.43	2.65	—
7.6	—	—	3.10	—
9.2	1.69	2.50	3.50	—
10.7	—	—	3.50	—
12.2	2.55	3.34	3.85	—
13.7	—	—	3.90	—
15.4	4.38	4.48	4.14	2.86
18.5	5.33	5.59	4.43	3.15
21.3	6.33	6.68	4.45	3.33
24.6	7.7	7.31	4.68	3.68
27.7	8.82	7.97	4.93	4.08
30.8	9.09	8.60	5.25	4.32
33.8	9.86	9.02	5.52	4.71
36.9	10.3	9.37	6.00	5.05
40.0	11.0	9.76	6.54	5.41
43.1	10.8	10.2	6.52	5.73
46.1	11.1	10.9	6.76	6.25
49.2	10.8	10.8	7.15	6.00
52.3	11.2	11.1	—	—
55.4	11.3	11.4	7.64	6.60
58.2	11.8	11.6	—	—
61.5	11.7	11.9	8.20	7.44
67.7	12.3	12.4	8.70	8.10
73.8	12.7	12.8	9.15	8.37
80.0	13.1	13.2	10.0	8.90
86.1	13.5	13.5	10.7	9.72
92.3	14.0	14.3	11.4	10.4

left axis corresponds to pure N₂ and the right axis to pure CO₂. Since N₂ does not form a stable negative ion, the data cannot be extended to the pure N₂ axis.

The drift velocity of electrons in these binary mixtures has been previously reported (Sierra *et al* 1979), and will not be discussed here. The data, however, are presented in table 2. It should be pointed out that at the highest E/N values (100 Td) the drift velocities reported here in pure CO₂ are about 10% lower than previously reported values (Lowke 1963). The agreement with Elford and Haddad (1980) is within $\pm 5\%$ in pure CO₂ in the region of overlap. Measurements in pure N₂ are also reported.

3.2. He:CO₂:N₂ mixtures

The role of helium in the CO₂ laser has been discussed by several authors (Nighan 1970, Patel *et al* 1965). Its principal effect is an increase in the electron energy over that observed in CO₂:N₂ mixtures at equivalent E/N . As a result, ionisation may be expected to be important in the ternary mixtures.

Figure 7 shows the observed *total* ion formation coefficient, $(\alpha + \eta)/N$, plotted against E/N for 12:4:1 and 8:1:1 mixtures. The solid lines are from the theoretical calculations of Lowke *et al* (1973). Above 4 Td in the 12:4:1 mixture we know from the theoretical calculations that $\eta < 0.1 \alpha$, and we are justified in comparing our experimental results

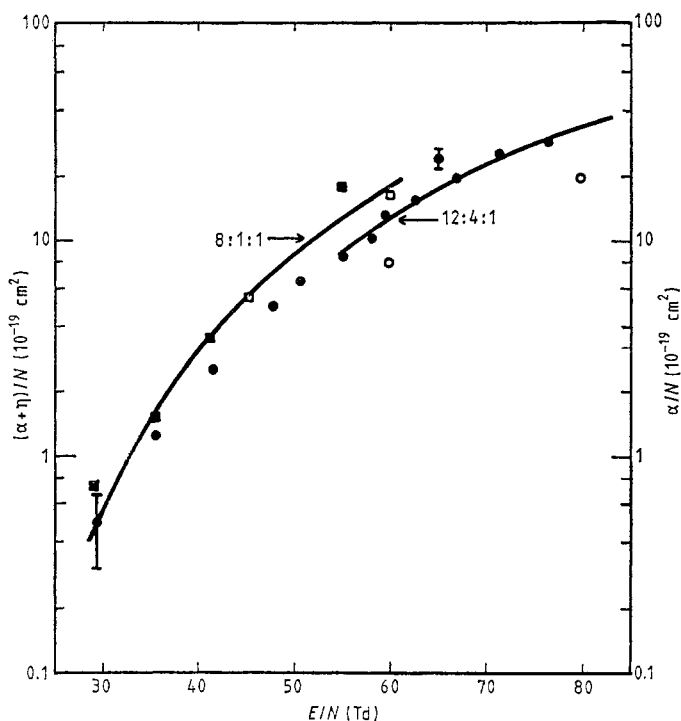


Figure 7. The left vertical axis shows $(\alpha + \eta)/N$ for the 8:1:1 mixture (■) and the right shows α/N for the 12:4:1 mixture (●). The full curve between 30 and 60 Td represents $(\alpha + \eta)/N$ as calculated by Lowke *et al* (1973) for the 8:1:1 case. The full curve between 55 and 85 Td is from the same authors and depicts α/N for the 12:4:1 mixture. The open circles and squares are the results of Lakshminarasimha *et al* (1976) for the 12:4:1 and 8:1:1 mixtures respectively, for $(\alpha - \eta)$.

with α/N . In the other case, (8:1:1), our experimental data are to be compared with calculated values of $(\alpha + \eta)/N$. Previously experimental cross-sections (Lakshminarasimha *et al* 1976, Haydon and McIntosh 1976) go down to values of about 10^{-18} cm². In the present work, values below 10^{-19} cm² have been measured. For the 8:1:1 case, there is excellent agreement between the present results and those of Lakshminarasimha *et al* (1976). The results of the latter investigators lie about 20% lower than the work reported here and theoretical calculations in the 12:4:1 mixture. Haydon and McIntosh have reported values for α/N in the 8:1:1 mixture above 60 Td which also show good agreement with the present results. Values of $(\alpha + \eta)/N$ for He:CO₂:N₂ are listed in table 3.

Table 3. Reduced total ion production coefficient $(\alpha + \eta)/N$ in He:CO₂:N₂ mixtures. E/N values are accurate to $\pm 3\%$. Results are accurate to $\pm 10\%$.

E/N (Td)	$(\alpha + \eta)/N$ (10^{-19} cm ²)	
	12:4:1	8:1:1
18.2	—	0.16
21.2	—	0.25
24.3	—	0.17
27.3	—	0.37
29.7	0.50	0.78
33.4	—	1.9
35.7	1.3	1.5
41.6	2.5	4.5
43.1	—	4.1
47.6	5.0	—
50.6	6.6	—
52.3	—	13
55.0	7.6	18
58.0	10.3	—
59.5	13.0	—
62.5	15.5	—
65.4	24.0	—
66.9	19.5	—
71.4	25.4	—
77.3	29.0	—
83.3	47.5	—

Recently, Sakai *et al* (1979) have carried out numerical calculations on the 8:1:1 mixture. Their results, over the E/N range reported here, are the same as Lowke *et al* (1973) at the lower end and a few per cent lower at the upper end.

The drift velocities of the electrons in both mixtures are presented in figures 8 and 9. The vertical bars represent the error limits imposed by the 10 ns resolution of the transient recorder at the largest drift velocities (shortest transit times), and the scatter in the data at the smaller drift velocities. Within the experimental accuracy there is little difference between the two sets of data. Figure 8 includes theoretical calculations by Leland (1979) and by Lowke *et al* (1973) for the 12:4:1 laser mixture. Above 10 Td the experimental points fall below both theoretical predictions. On the other hand, in the 8:1:1 mixture, the agreement between theory and experiment is excellent. For reference purposes, the drift velocity results are printed in table 4.

Many experimental factors place a limit on the absolute accuracy of the results presented in this paper. However, precautions were taken to ensure the most precise

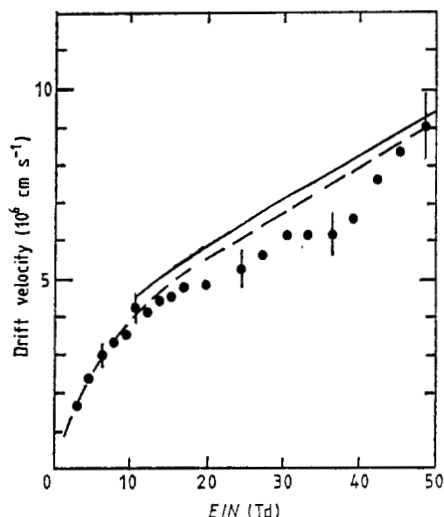


Figure 8. Electron drift velocity in He:CO₂:N₂=12:4:1. ●, experimental results. The full and broken curves are due to Leland (1979) and Lowke *et al* (1973), respectively.

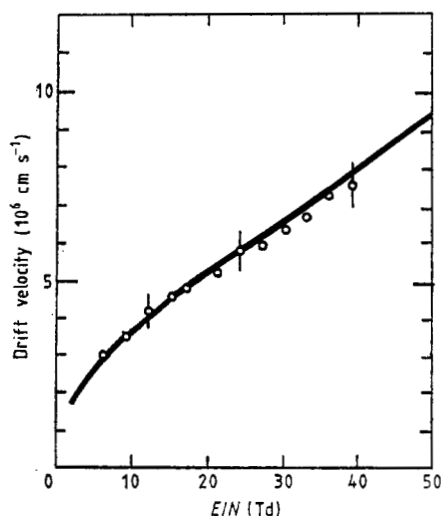


Figure 9. Electron drift velocity in He:CO₂:N₂=8:1:1. ○, experimental results. The theoretical results of Leland (1979) and of Lowke *et al* (1973) fall within the thickness of the full curve.

measurements possible were obtained. The distance between the electrodes, the applied electric field, and the pressures of the gases used in the experiment were all known to within $\pm 1\%$, placing a limit on the E/N values of $\pm 3\%$. Since the measured coefficients α/N and η/N are so small, the greatest source of error would be gas impurities that have much larger ionisation and attachment coefficients. The gases used in this experiment limited the total impurity concentration to less than 1 part in 100 000 which requires reduced ion formation coefficients greater than 10^{-14} cm² to change the results obtained by 1%. Over the E/N range studied in this experiment, none of the impurities, known to

Table 4. Drift velocity of electrons in He:CO₂:N₂ mixtures. E/N values are accurate to $\pm 3\%$. Results are accurate to $\pm 5\%$.

E/N (Td)	W (10^{-6} cm s $^{-1}$)	
	12:4:1	8:1:1
3.0	1.64	—
4.5	2.32	—
6.0	2.99	2.99
7.6	3.33	—
9.1	3.49	3.42
10.6	4.25	—
12.1	4.08	4.02
13.6	4.35	—
15.1	4.55	4.50
16.5	4.78	—
18.1	4.88	4.87
19.6	4.88	—
21.1	5.55	5.23
24.2	5.28	5.80
27.2	5.71	5.88
30.2	6.25	6.29
33.0	6.25	6.76
36.1	6.25	7.14
39.1	6.70	7.41
42.1	7.69	—
45.1	8.39	—
48.1	9.09	—

be present, have such large ion formation coefficients. Therefore all possible experimental errors have less influence on the results than the scatter in the data.

4. Summary and conclusions

In this work, data are presented for some of the more important swarm parameters, the attachment probability per unit length η , Townsend's ionisation coefficient α , and the drift velocity of electrons. The results are compared with several numerical studies and previous data. Overall agreement with previous work is good, the previous ionisation data of Lakshminarasimha *et al* (1976) for the 12:4:1 case lie about 20% lower than the work reported here. In general, the validity of the theoretical method and input cross-sections is supported by the present work.

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