MEASUREMENTS OF TOWNSEND'S ENERGY FACTOR k_1 FOR ELECTRONS IN CARBON DIOXIDE

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[*Manuscript received July* 20, 1964]

Summary

Values of Townsend's energy factor k_1 for electrons in carbon dioxide have been determined as a function of the parameter E/p for $0 \cdot 1 \leqslant E/p \leqslant 50$ at a temperature of 293°K. The results are compared with those of other investigators and are used in a recomparison of the cross sections for electron attachment deduced from swarm and beam types of experiments for swarms of electrons having mean energies of up to 5 eV.

I. Introduction

When swarms of electrons, having mean energies of the order of a few electron volts, pass through electronegative gases, the possibility exists that the electrons will interact with the gas atoms to form negative ions by electron attachment. In addition, the electrons may ionize the gas atoms through the so-called Townsend primary ionization process. For a full understanding of the mechanisms controlling the processes of attachment and ionization in a particular gas, it is necessary to have available reliable data on such parameters as the mean energy, drift velocity, and velocity distribution of the electrons.

For the particular case of carbon dioxide, much of the available data is conflicting, especially that for Townsend's energy factor k_1 (or its equivalent, D/μ) which estimates the mean energy of the swarm of electrons. The present paper gives details of a redetermination of k_1 for electrons in carbon dioxide over a wide range of values of the parameter E/p (where E = electric field in V/cm and p = gas pressure in torr) and compares the results obtained with those of earlier investigators. In addition, the values found for k_1 are utilized in a re-evaluation of the available data on the attachment of electrons in carbon dioxide.

II. Experimental Procedure

The apparatus used in the present investigation has been fully discussed by Crompton and Jory (1962), and was based on Huxley's modification of Townsend's original method of analysing the lateral diffusion of a stream of electrons (Huxley 1940). A schematic diagram of the apparatus is shown in Figure 1. Electrons generated by the heated filament F enter the diffusion chamber through the hole (of 1 mm diameter) in the cathode C and drift under the influence of the applied electric field to the receiving electrode A. The anode A comprises a central disk and surrounding annuli.

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It has been shown by Huxley (1959) that in the case of a stream of electrons diffusing through a gas in which both ionization and attachment processes are occurring, the ratio R of the current received by the annulus A_2 to the sum of the currents received by the annuli A_2 and A_3 is given by

$$R = 1 - \frac{\frac{h}{d_b}\exp(\lambda h-\mu d_b)+\frac{\lambda h\alpha_a}{\mu}\int_0^1 \exp(\lambda hs)\left\{\exp\left[-\mu h\left(\frac{b^2}{h^2}+s^2\right)^{\frac{1}{2}}\right]-\exp\left[-\mu h\left\{\frac{b^2}{h^2}+(2-s)^2\right\}^{\frac{1}{2}}\right]\right\}\mathrm{d}s}{\frac{h}{d_a}\exp(\lambda h-\mu d_a)+\frac{\lambda h\alpha_a}{\mu}\int_0^1 \exp(\lambda hs)\left\{\exp\left[-\mu h\left(\frac{a^2}{h^2}+s^2\right)^{\frac{1}{2}}\right]-\exp\left[-\mu h\left\{\frac{a^2}{h^2}+(2-s)^2\right\}^{\frac{1}{2}}\right]\right\}\mathrm{d}s}, \quad (1)$$

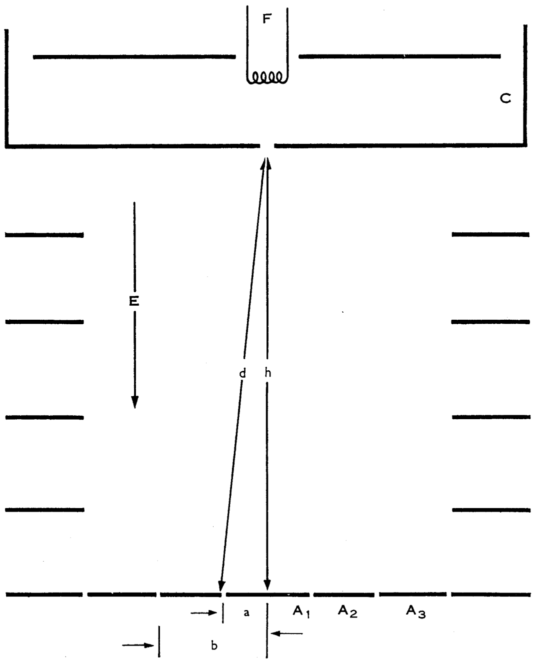

Fig. 1.—Schematic diagram of diffusion chamber.

where h = length of the diffusion space,

$d_b = \sqrt{(b^2+h^2)}$,

$d_a = \sqrt{(a^2+h^2)}$, where a and b are the inner and outer radii of the inner annulus,

$2\lambda = W/D = \dfrac{\text{drift velocity of electrons}}{\text{diffusion coefficient of electrons}} = 39\cdot 79\ E/k_1$ at 20°C,

k_1 = Townsend's energy factor,

$\mu^2 = \lambda^2+2\lambda\alpha$,

$\alpha = \alpha_a-\alpha_i$ = (attachment coefficient)—(ionization coefficient).

When α_a is small, equation (1) reduces to

$$R = 1-(d_a/d_b)\exp\{\mu(d_a-d_b)\} \tag{2}$$

and when $\alpha_i = \alpha_a = 0$, equation (1) reduces to the well-known form

$$R = 1-(d_a/d_b)\exp\{\lambda(d_a-d_b)\}. \tag{3}$$

Measurements of the ratio R were made for a temperature of 293°K at a number of values of $h(h = 3, 6, 10$ cm) and of $p(p = 1, 2, 4, 8$ torr) over a wide range of values of the parameter E/p $(0\cdot1 \leqslant E/p \leqslant 50)$. Wherever possible, the experimental parameters were chosen so that any negative ions formed between the filament F and the cathode C (Fig. 1) and passing into the diffusion region CA, were collected on the central disk A_1 which was kept at earth potential throughout the experiment.

The gas used in the investigation was carbon dioxide of commercial purity. Traces of the permanent gases were removed by repeatedly freezing the carbon dioxide (using liquid air) and pumping on the frozen carbon dioxide for extended periods. To remove any traces of water vapour or other readily condensable impurities, the gas was admitted to the diffusion apparatus slowly through copper-filled traps surrounded by a mixture of solid carbon dioxide and acetone. A drying tube of phosphorus pentoxide was connected permanently to the gas reservoir. The diffusion apparatus could not be baked but the background degassing rate of the system was of the order of 10^{-5} torr/hr so that contamination of the gas samples used from this source was negligible during the course of a set of measurements. Measurements for $E/p \leqslant 10$ were also taken using Airco "Reagent Grade" carbon dioxide, with no change in the results.

III. Results

The results obtained for k_1 are summarized in Tables 1 and 2 and are shown in Figures 2 and 3. To facilitate discussion, the results may be divided into three groups, according to the range of values of E/p in which they were obtained.

(a) $5 < E/p < 20$

For this range of values of E/p, the use of equation (1) in conjunction with an extrapolation of reported values of α_a/p showed that electron attachment was unlikely to affect the current ratios appreciably at any combination of h and p used in the investigation. For this reason the use of the simpler equations (2) or (3) to analyse the results, appeared to be justified. When this procedure was adopted, using an extrapolation of the values of $\alpha = (\alpha_a-\alpha_i)$ given by Bhalla and Craggs (1960), which are in good agreement with those of Schlumbohm (1962), the results obtained for k_1 showed no dependence on either h or p, thus confirming that the influence of attachment was negligible. Furthermore, the results obtained using equation (2) were identical with those obtained using equation (3) showing that the influence of ionization was also negligible.

(b) $20 < E/p \leqslant 50$

Since values of k_1 for high values of E/p are required for use in conjunction with determinations of ionization and attachment coefficients, an attempt was made in the present investigation to take measurements of k_1 for values of E/p which were as high as possible. The upper limit of $E/p = 50$ was fixed by the onset of electrical breakdown of the gas in the apparatus.

For $E/p > 20$, the use of equations (2) or (3) for the analysis of the measurements was again justified for the smallest values of h and p used. However, in this range of E/p the results obtained using equation (2) differed slightly from those obtained using equation (3), the discrepancy between the two sets of data amounting to a maximum of 2% at $E/p = 50$. This discrepancy represents the influence of ionization on measurements made under experimental conditions for which the effects of attachment were negligible. The results obtained using both equations are shown in Table 1, the accuracy of the experimental measurements being about $\pm 1\%$. It is perhaps worth emphasizing that the use of equation (3) for the analysis of measurements made for any gas, at values of E/p for which α_i/p is appreciable, leads to values of k_1 which, although showing no dependence on the geometry of the apparatus or on the gas pressure used, are nevertheless higher than the true values. The true values of k_1 can only be obtained using equation (2) in conjunction with known values of α_i/p.

TABLE 1

VARIATION OF k_1 WITH E/p FOR $6 \leqslant E/p \leqslant 50$ AT 293°K

E/p (Vcm^{-1} $torr^{-1}$)	k_1 from eqn. (3)	k_1 from eqn. (2)	D/μ from eqn. (2)	E/p (Vcm^{-1} $torr^{-1}$)	k_1 from eqn. (3)	k_1 from eqn. (2)	D/μ from eqn. (2)
6·0	13·4	13·4	0·338	18·0	72·7	72·7	1·84
7·0	20·2	20·2	0·510	20·0	78·9	78·9	1·99
8·0	27·6	27·6	0·697	25·0	95·0	95·0	2·40
9·0	33·8	33·8	0·854	30·0	108·4	108·4	2·74
10·0	40·1	40·1	1·01	35·0	120·3	120	3·03
12·0	50·3	50·3	1·27	40·0	130·9	130	3·28
14·0	58·6	58·6	1·48	45·0	140·7	139	3·51
16·0	66·0	66·0	1·67	50·0	148·7	146	3·69

At the highest values of h and/or p used in the present investigation, the measurements taken for $E/p > 20$ were noticeably affected by attachment and it was necessary to use the complete equation (1) for their analysis. The use of equation (1) necessitated the employment of values of α_a/p and α_i/p and since the available data for these quantities are rather conflicting, the analysis was carried out using firstly Bhalla and Craggs's data, and secondly Schlumbohm's data. Using Bhalla and Craggs's data, the values of k_1 were up to 9% higher than those given in Table 1 (column 3), while using Schlumbohm's data the discrepancies were up to 4%. Until more reliable data for α_a/p become available (the accuracy claimed by Schlumbohm for his measurements of this quantity is $\pm 25\%$) it is not possible to determine accurately what correction to the results of Table 1 for $E/p > 20$ is necessary. It is not expected, however, that the correction will be more than about 5% at $E/p = 50$ (decreasing rapidly with decreasing E/p) and may well be less than this.

(c) $0 \cdot 1 \leqslant E/p < 5$

Preliminary measurements showed that over a considerable portion of this range of E/p, the values of k_1 for electrons were likely to be small, and that, consequently, the current distribution at the receiving electrode for both ions and electrons entering the diffusion chamber through the source hole would be similar. Hence, if any negative ions were formed by attachment in the region FC (Fig. 1) it would become difficult, if not impossible, to arrange the experimental conditions to ensure that the current ratios were not falsified by the presence of an unknown fraction of ions entering the chamber with the electrons.

For this reason, measurements of k_1 for $E/p \leqslant 5$ were taken in a second apparatus in which the electron stream passed through a Bradbury and Nielsen drift-velocity apparatus before entering the diffusion chamber. It was expected that a suitable choice of the experimental parameters would enable any negative ions present in the electron stream to be removed in the drift-velocity section of the apparatus, thus ensuring that the values of k_1 determined in the diffusion chamber were not falsified. However, it was found that the number of negative ions in the electron stream was in fact negligibly small so that no "tuning" of the drift-velocity apparatus was necessary. The values of k_1 measured in the diffusion chamber of the auxiliary apparatus were in good agreement with those measured in the main apparatus. Since no evidence of negative ions could be found at values of $E/p \leqslant 5$, a final set of measurements was taken in the main apparatus, using it in the more conventional manner, that is, by measuring the ratio of the current to the central disk A_1 (Fig. 1) to that to the annular rings A_2+A_3. The results found in this way for k_1 were entirely self-consistent and agreed with measurements taken using the apparatus as described in Section II above. Table 2 summarizes the results obtained, the values of k_1 being accurate to $\pm 1\%$.

Table 2

Variation of k_1 with E/p for $0 \cdot 1 \leqslant E/p \leqslant 5$ at 293°K

E/p ($\text{Vcm}^{-1}\text{ torr}^{-1}$)	k_1 from eqn. (2) or (3)	D/μ	E/p ($\text{Vcm}^{-1}\text{ torr}^{-1}$)	k_1 from eqn. (2) or (3)	D/μ
0·100	1·01	0·0255	1·50	1·34	0·0338
0·200	1·02	0·0258	2·00	1·53	0·0386
0·400	1·05	0·0265	3·00	2·25	0·0568
0·600	1·09	0·0275	4·00	3·75	0·0947
0·800	1·13	0·0286	5·00	7·45	0·188
1·000	1·19	0·0301			

IV. Discussion of Results

It is of interest to compare the results given in Tables 1 and 2 with those of previous investigations. It may be seen from Figure 2 that at high values of E/p (> 5) there is general agreement between the present results and those of Skinker (1922), Bailey and Rudd (1932), and Warren and Parker (1962), the discrepancies being less than the probable combined experimental errors. At the highest values of E/p, the present results are approximately 10% higher than those of Skinker, which are the only other data in this range. Discrepancies of this order of magnitude can be significant in calculations of ionization and attachment cross sections (see Section V).

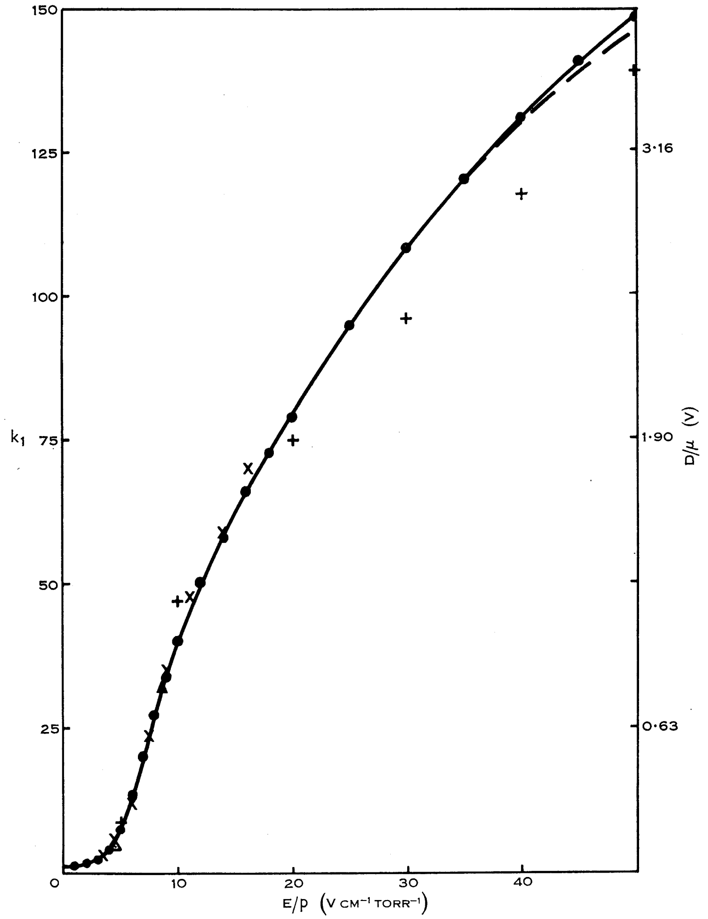

Fig. 2.—Variation of k_1 with E/p for $E/p \leqslant 50$ at 293°K. ● Present experimental points. Broken line shows values of k_1 corrected for the influence of ionization. △ Warren and Parker (1962). × Bailey and Rudd (1932). + Skinker (1922).

For low values of E/p (< 5), Figure 3 demonstrates the large differences which exist between the results of the various investigations. The present results are in good agreement with those of Warren and Parker, both sets of data lying well below those of Skinker and of Cochran and Forester (1962). The recent results of Hurst *et al.* (1963) have been omitted from Figure 3 because of the large experimental scatter they exhibit. The results of Bailey and Rudd are in reasonably good agreement with the present results and those of Warren and Parker, with the exception of their value of k_1 at $E/p = 2$. It seems probable that the results of Cochran and Forester are incorrect, since it appears unlikely that their data would extrapolate to give $k_1 = 1$ at $E/p = 0$. To a lesser extent, the same is true of Skinker's data.

The maximum discrepancy between the present results and those of Warren and Parker is about 5%. In favour of the present results it should be noted that while the values of k_1 given in Tables 1 and 2 may be extrapolated smoothly to $k_1 = 1$ at $E/p = 0$, the measurements of Warren and Parker tend to a limit of k_1 which is slightly less than one.

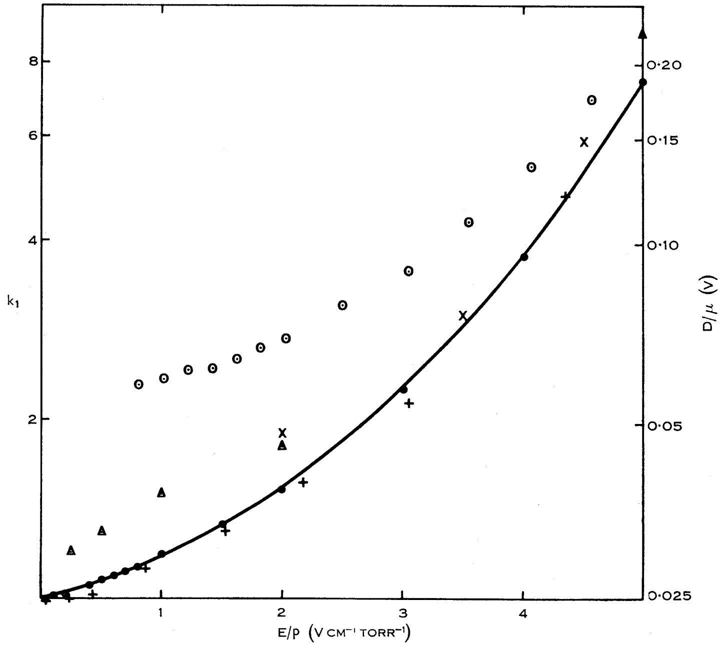

Fig. 3.—Variation of k_1 with E/p for $E/p \leqslant 5$ at 293°K.
● Present data. + Warren and Parker (1962).
× Bailey and Rudd (1932). ⊙ Cochran and Forester (1962).
△ Skinker (1922).

V. Attachment Cross Sections

Bhalla and Craggs (1960) deduced from their measurements of attachment and ionization coefficients the cross sections for ionization and attachment by swarms of electrons having mean energies $\bar{\epsilon}$ in the range $0 < \bar{\epsilon} < 5$ eV. These cross sections were compared with those deduced from the beam experiments of Craggs and Tozer (1960), the calculations being carried out for Maxwellian and Druyvesteyn distributions of velocities. More recently swarm measurements of ionization and attachment coefficients have been conducted by Schlumbohm (1962), while further beam experiments have been carried out by Schulz (1962) and by Asundi, Craggs, and Kurepa (1963). The agreement between the results obtained by Schulz and by Asundi, Craggs, and Kurepa is good, both for the electron energies at which the cross sections are at a maximum and for the magnitudes of these maxima. Recently, Asundi and Craggs (1963) have recompared the results of the various investigations.

When comparisons are made between the results of swarm and beam experiments, values of Townsend's energy factor k_1 and of the drift velocity W are employed as auxiliary data, and the accuracy of the comparisons depends markedly on the accuracy of the values used for k_1 (Rees and Jory 1964) and to a lesser extent on those used for W. The sensitivity of the comparisons to the accuracy of the data used for k_1, arises from the fact that, particularly if k_1 increases slowly with E/p, a small error in k_1 can lead to a large error in the value of E/p (and hence also in α_a/p) attributed to a given mean energy $\bar{\epsilon}$. It was thought to be worth while in the present work

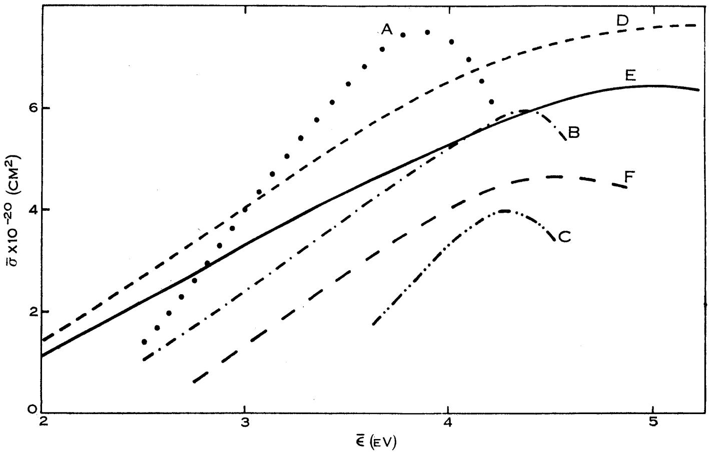

Fig. 4.—Variation of mean cross section $\bar{\sigma}$ for attachment of electrons with mean energy $\bar{\epsilon}$, assuming a Druyvesteyn distribution of velocities.
Curve A, swarm data of Bhalla and Craggs (1960) using Skinker's values of k_1 and W.
Curve B, curve A recalculated using k_1 data of Table 1 and W data of Table 3.
Curve C, swarm data of Schlumbohm (1962) using data of Tables 1 and 3.
Curve D, beam data of Asundi, Craggs, and Kurepa (1963).
Curve E, beam data of Schulz (1962).
Curve F, beam data of Craggs and Tozer (1960).

Table 3

VALUES OF DRIFT VELOCITY W USED IN CROSS-SECTION CALCULATIONS

E/p (Vcm^{-1} torr^{-1})	20	25	30	35	40	45	50
$W \times 10^{-5}$ (cm/s)	124	126	127	129	139	152	166

to recalculate the cross sections for attachment from the swarm data of Bhalla and Craggs and of Schlumbohm using the values of k_1 given in Table 1, column 3, in place of those of Skinker, which were the only ones previously available. In addition, the values of W shown in Table 3 were used. At high E/p the values of W determined recently by Frommhold (1960) were used in place of Skinker's earlier values, which had been determined using a more indirect method. For lower E/p the values of W were obtained by extrapolating Frommhold's data to merge with the data obtained at low E/p by Pack, Voshall, and Phelps (1962). The recalculated cross sections are shown in Figure 4 for the case of a Druyvesteyn distribution of electron energies, together with the cross sections obtained by suitably averaging the available beam data. The corresponding results for a Maxwellian distribution of energies are shown in Figure 5.

It is seen from Figure 4 that some measure of agreement can be obtained between the results of swarm and beam experiments for a Druyvesteyn distribution. The agreement is better than that obtained in Bhalla and Craggs's work and in that of Asundi, Craggs, and Kurepa which, of necessity, utilized the data for k_1 and W obtained by Skinker. The effect of small errors in k_1 on the attachment cross sections

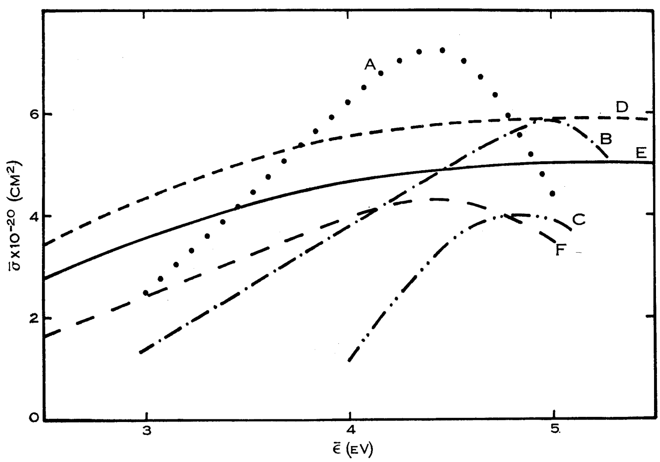

Fig. 5.—As for Figure 4, assuming a Maxwellian velocity distribution.

is striking, an error of 5% in k_1 at $E/p = 30$ producing an error of approximately 40% in the corresponding cross section. Another noteworthy feature of Figure 4 is that, in spite of the good agreement between the results of the beam experiments of Schulz and of Asundi, Craggs, and Kurepa, such discrepancies as do exist are sufficient to give mean cross sections $\bar{\sigma}$ which differ by up to 20%. It is seen from Figure 5 that the agreement existing for a Maxwellian distribution is poorer than that for a Druyvesteyn distribution.

The present calculations and those of Asundi and Craggs illustrate the marked influence of relatively small errors in k_1 on the calculations of cross sections. It appears also that a Druyvesteyn velocity distribution is a better approximation to the velocity distribution than is a Maxwellian distribution. However, to proceed further more accurate measurements of W and of α_a/p are required.

VI. Conclusions

Values of k_1 for electrons in carbon dioxide have been determined at a temperature of 293°K for a wide range of E/p ($0 \cdot 1 \leqslant E/p \leqslant 50$). For the highest values of E/p (> 20) the results differ by up to 10% from those of Skinker. At intermediate values of E/p ($5 < E/p < 20$) the results agree with those of other workers, while for $E/p < 5$ the results confirm the general trend of the results of Warren and Parker, and of Bailey and Rudd, and indicate that the results of Skinker and of Cochran and Forester are considerably too high in this region of E/p.

An analysis of measurements made at $E/p \geqslant 25$ under conditions for which electron attachment and ionization were significant has shown the ionization and attachment coefficients measured by Schlumbohm to be rather more consistent with the present results than those measured by Bhalla and Craggs.

A re-evaluation of the cross sections for electron attachment in carbon dioxide has been carried out using the present values of k_1. The calculations show a Druyvesteyn distribution of velocities to give some measure of agreement between the results of swarm and beam experiments, and emphasize the need for using data of considerable accuracy if the comparisons are to be worthwhile. More accurate determinations of W and α_a/p for electrons in carbon dioxide are therefore required for use in conjunction with other accurate data now available.

VII. Acknowledgments

The author is greatly indebted to Professor Sir Leonard Huxley, Dr. R. W. Crompton, and the other members of the Ion Diffusion Unit for their willing advice and assistance at all times.

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