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Ionization of gases by a pulsed electron beam as studied by self-focusing. III. He, Ar, and O_2 mixtures

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The self-focusing of a pulsed electron beam from a Febetron 706 in He, Ar, and O_2 is enhanced by 10% addition of polyatomic gases at total pressure higher than 50 Torr. The enhancement by addition of N_2 to He is stronger than that expected from pure N_2. The effect of additives to Ar appears abruptly from a certain pressure dependent on additives. According to the conclusion in part II, this enhancement is caused by the increasing of the mean ionization time t_i of secondary ionization due to electric field E induced by the pulsed beam. Such inference is demonstrated in the present paper on the basis of the time t_i for a mixture given by Eq. (1); t_i for a mixture is determined by the ionization potential of additives relative to that of a host gas, the initial slope of the energy dependence of the total ionization cross section σ_i of additives, and the retardation of secondary electrons by additives for acceleration by E. When a gas having small σ_i and large effect of slowing down is added, the self-focusing is strongly enhanced due to the large increase of t_i. This increase of t_i is also demonstrated by the increasing of $1/(w\alpha)$ by addition of additives (w is the electron drift velocity and α is the first Townsend ionization coefficient); the values of both parameters for some cases can be known either from papers of experimental data and the Boltzmann equation analysis or by the average electron method.

INTRODUCTION

As described in detail in Part II,[1] the dose D_{obs} on the beam axis for a pulsed electron beam from a Febetron 706 increases gradually with increasing pressure p after passing a minimum; usually, D_{obs} becomes the minimum at 5–10 Torr of polyatomic gases. The value of D_{obs} is relatively small in He, Ar, and O_2 as compared with other gases.[2] When polyatomic gases are added to those gases, the increase of D_{obs} with pressure is enhanced so that the self-focusing of the beam is enhanced by addition of additives.[3] In a previous paper,[3] the effect of additives has been discussed on the basis of the promotion of electron–ion recombination processes due to the slowing down of secondary electrons by additives and of the occurrence of charge transfer processes to additive molecules. However, this is rather inconsistent with the conclusion in Part II[1] that these processes can be neglected for the beam with ns pulse duration. Penning ionization processes are also almost negligible in this case because of their slow rate constants.

According to the conclusion in Part II,[1] such an effect of additives indicates that the mean ionization time t_i of secondary ionization due to electric field E_z induced by the pulsed beam is lengthened by additives. In the present paper, the previous data for the mixtures[3] will be interpreted in terms of t_i in the mixtures. In the following analysis, E/p represents E_z/p.

ESTIMATION OF t_i IN A BINARY MIXTURE

When there are no effective electron attachment and Penning ionization processes, we can assume the equation of t_i in a binary mixture as

$$\frac{1}{n_0 t_i} = \left(\frac{2}{m_e}\right)^{1/2} \left[\int_{IP_1}^{\infty} x \, \epsilon^{1/2} \sigma_{i1}(\epsilon) F(\epsilon) \, d\epsilon\right.$$

$$\left. + \int_{IP_2}^{\infty} (1-x) \, \epsilon^{1/2} \sigma_{i2}(\epsilon) F(\epsilon) \, d\epsilon\right] , \qquad (1)$$

where n_0 is the total number density of gas molecules proportional to total pressure p, m_e the electron rest mass, x the mole fraction of a host gas, IP the ionization potential, and $F(\epsilon)$ the energy distribution function of secondary electrons. The indexes 1 and 2 are referred to a host gas and an additive, respectively. Yamane[4] has interpreted the first Townsend ionization coefficient α in Ar mixtures in terms of the similar parameters; the initial slope of energy dependence of σ_1, IP of additives relative to that of a host gas, the average fractional energy loss per collision, and the mean energy of secondary electrons $(\bar{\epsilon})$.

Data on σ_i and IP for lower energy electrons have been measured for several gases by Rapp and Englander-Golden[5,6] as shown in Fig. 1. As has been described in Part II,[1] the $Q_t f - \bar{\epsilon}$ curve is useful as a measure of $F(\epsilon)$, where Q_t is the total collision cross section and f the mean fractional energy loss per collision. Therefore, we can interpret the data on D_{obs} in the present mixtures qualitatively in terms of the lengthened t_i estimated by Eq. (1) in referring to data on these parameters.

In Eq. (1), the first integral represents the contribution of a host gas to t_i. When this integral is very small compared to the second integral for an additive, t_i in the mixture is mainly determined by the properties of the additive; in the other words, D_{obs} in the mixture can be estimated from D_{obs} in the pure additive. Since He has the highest IP and the small σ_i, the contribution of He to t_i for He mixtures becomes easily negligibly small by addition of polyatomic gases which make $F(\epsilon)$ shift to the lower-ϵ side. Since He has the small σ_i, secondary electrons generated from He hardly contribute to the
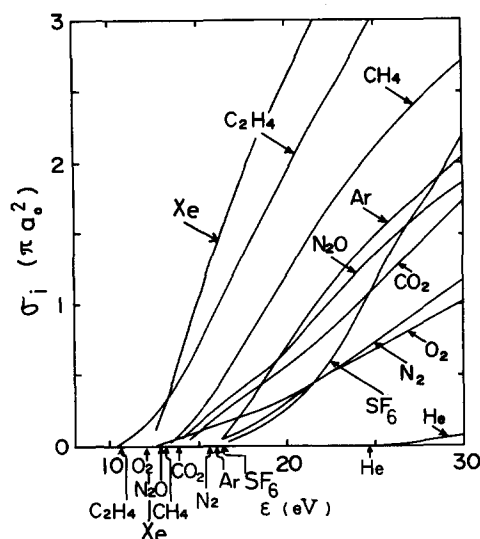
FIG. 1. Total ionization cross sections of various gases by electron collision in πa_0^2 unit as functions of electron energy, cited from the data by D. Rapp and P. Englander-Golden. An arrow indicates an ionization potential.

second integral. Therefore, D_{obs} in He mixtures should be generally estimated by D_{obs} in the pure additive. This is approximately realized in the present data for He mixtures except N_2 as described later. In Fig. 1, the situation is not so simple for Ar and O_2 as for He.

According to data on D_L/μ,[7] $\bar{\epsilon}$ in Ar is supposed not to be reduced from 9 eV down to E/p of a few V cm^{-1} $Torr^{-1}$. Therefore, secondary electrons in the pulsed-beam channel are not retarded in Ar so that they have plenty of chances for ionization. Their chances to Ar are lost only by their retardation by additive molecules. In fact, the effect of the enhancement is weak for Ar mixtures with a weakly retarding gas.

The discussion on the basis of Eq. (1) should be realized macroscopically in swarm parameters. The equation of t_i can be rewritten as

$$1/(pt_i) = w_m \alpha_m/p \quad \text{for } \eta_m = 0 , \tag{2}$$

where w is the electron drift velocity, η the electron attachement coefficient, and m refers to mixture. Data on w_m and α_m/p for some mixtures have been determined either experimentally or by the Boltzmann equation analysis. On the other hand, as a first approximation, the average electron method has been proposed to estimate w_m in a binary mixture.[8] When values of w and ϵ_k (characteristic energy) are known as functions of E/p for two gases, 1 and 2, respectively, the value of w_m at $E/p = z_m$ is given by

$$w_m z_m = xw_1 z_1 + (1 - x)w_2 z_2 \tag{3}$$

and

$$z_m = xz_1 + (1 - x) z_2 , \tag{4}$$

where z_1 and z_2 are the values of E/p giving the same ϵ_k for respective gases and w_1 and w_2 are the values at these z's for these gases. The value of α_m/p at $E/p = z_m$ is similarly estimated by

$$(\alpha_m/p) = x (\alpha_1/p) + (1 - x) (\alpha_2/p) , \tag{5}$$

where α_1 and α_2 are the values at z_1 and z_2 at which $\epsilon_k = \epsilon_m$, respectively. For the present estimation, it is assumed that

$$\bar{\epsilon} = \tfrac{3}{2} \epsilon_k = \tfrac{3}{2} e(D_L/\mu) . \tag{6}$$

Related data for Eqs. (3)–(6) are cited from references in Part I[9] and II.[1]

Values of pt_i given by $p/(w_m \alpha_m)$ from these methods are in a function of not p but E/p. The value increases with decreasing E/p which decreases with increasing p and also with lapse of time even at the same pressure.[1] Therefore, since D_{obs} is integrated over the whole pulse duration, D_{obs} increases more effectively with increasing pressure than pt_i with decreasing E/p. As compared at the same pressure, the increment of D_{obs} for a mixture from a pure host gas becomes larger than pt_i, shown as a function of E/p. Furthermore, the $D_{obs} - t$ curve becomes steeper than that expected from the $pt_i - E/p$ curve due to the variation of the beam emittance with the self-focusing.[9]

In conclusion, it should be noticed that the parameters for t_i are in functions of E/p for which p is not the partial pressure of an additive but the total pressure even if some correction is needed for it. The effect of some additive (for example, SF_6 in Ar) is found from lower pressure at which the effect cannot be expected from data on D_{obs} at the partial pressure of the pure gas. Such an effect can be explained by the above interpretation for E/p.

RESULTS AND DISCUSSION

The experimental methods have been described in detail in previous papers.[2,3,10] Mixtures were prepared in advance in a large reservoir for each run.

A. He mixtures

Values of D_{obs} for He mixtures containing N_2, O_2, N_2O, CH_4, C_2H_4, and SF_6 ($x = 0.9$) are shown as functions of total pressure in Fig. 2. The value increases gradually from 50 Torr by addition of these additives. The enhancement of the self-focusing is in the order

$$N_2 > N_2O, \quad C_2H_4, \quad SF_6 > O_2, \quad CH_4 . \tag{7}$$

When this order is compared with the order of D_{obs} in pure gases (Fig. 7 of Ref. 2), the self-focusing is remarkably enhanced by addition of N_2. The relationship of D_{obs} among other gases is almost the same between the pure gases and the mixtures, though data in the mixtures are rather scattered. This fact indicates that the effect of these additives except N_2 can be interpreted in terms of the properties of the additives as pointed out in Part II[1] because, in Eq. (1), the first integral for He is much smaller than the second integral for the additive. The curve of D_{obs} for pure C_2H_4, which is not shown in Ref. 2, is almost the same as that for CH_4,[11] while the curve for C_2H_4/He in Fig. 2 is rather higher than for CH_4/He. This may be due to reduction of $\bar{\epsilon}$ by C_2H_4 more effectively than by CH_4.[7] Decline of D_{obs} with increasing pressure above 250 Torr is mainly attributed
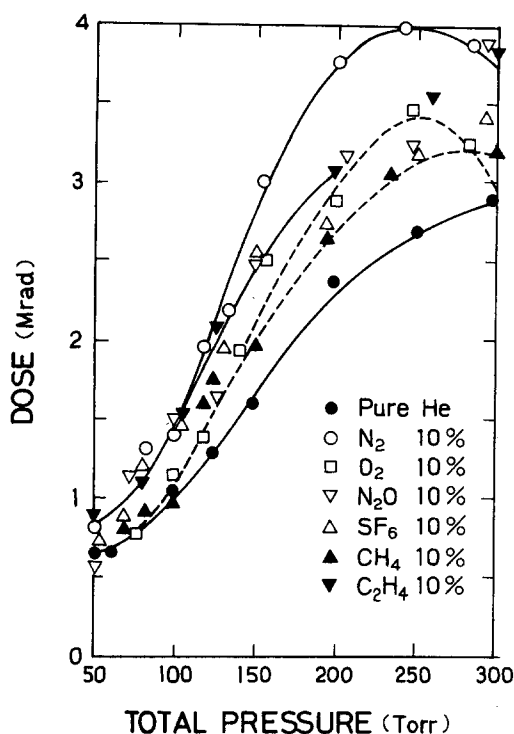
FIG. 2. The dose observed on the beam axis in pure He and He mixtures as functions of total pressure; ● pure He, ○ N_2/He (1/9), □ O_2/He (1/9), ▽ N_2O/He (1/9), △ SF_6/He (1/9), ▲ CH_4/He (1/9), and ▼ C_2H_4/He (1/9).

to multiple scattering of the primary beam by a background gas.

In Fig. 2, the increasing D_{obs} is strongly enhanced by addition of N_2 as shown as the order in Eq. (7). In Fig. 1, N_2 has the high IP and the small σ_i as compared with other additives. Furthermore, N_2 makes $\bar{\epsilon}$ reduce effectively.[1] Then, in Eq. (1), the value of the second integral for N_2 is also small as is the first integral for He so that the value of t_i is much lengthened due to these properties of N_2. This fact indicates that the self-focusing in He is strongly enhanced by addition of a gas having high IP, small σ_i, and the strong effect on $F(\epsilon)$. Haydon and McIntosh[12] have reported that α_m/p for N_2/He increases by about 10% between 17 and 400 V cm^{-1} $Torr^{-1}$. Although their data seem to be inconsistent with the above discussion, their data contain the contribution of the Penning ionization which can be ignored for the present study.

We attempted to estimate values of w_m and α_m/p by means of Eqs. (3)–(5) from data in pure gases already cited in Parts I[9] and II.[1] As compared with pure He, values of w_m decrease considerably for N_2/He (1/9) and CH_4/He (1/9), while the value of α_m/p for CH_4/He increases appreciably and that for N_2/He decreases only a little. Then, values of $p/(w_m \alpha_m)$ vary with E/p as shown in Fig. 3. The curves indicate that, from Eq. (2) the value of pt_i increases by addition of N_2 while the value is hardly affected by CH_4. When the pt_i–E/p curve is replotted as the D_{obs}–p curve, as described in the foregoing section, the latter curve for a mixture should be more separated from that for a pure gas than the former curve. Therefore, the curves of pt_i estimated by means

of the average electron method represent fairly well the aspect of the D_{obs} curves for these He mixtures.

According to the Boltzmann equation analysis with the assumption of no Penning ionization for SF_6/He (1/9),[13] the value of w_m at E/p of 100 V cm^{-1} $Torr^{-1}$ is smaller by 25% than for pure He and the value of α_m/p at the same E/p is larger by 35% than for pure He. Morruzi and Craggs[14] have reported that the value of α_m/p for SF_6/He (13/87) decreases by about 20% at E/p between 14 and 32 V cm^{-1} $Torr^{-1}$. These data suggest that the value of pt_i, given by $p/(w_m \alpha_m)$, in the mixture is almost the same as that in pure He at E/p of 100 V cm^{-1} $Torr^{-1}$, while this value becomes much larger at lower E/p than that in pure He. On the other hand, in Fig. 10 of Part I,[9] E/p in pure He is less than 100 V cm^{-1} $Torr^{-1}$ for the major part of the pulse duration at pressures higher than 50 Torr. Then, the effect of SF_6 in Fig. 2 is attributed to such a lengthened t_i by addition of SF_6. The value of $\bar{\epsilon}$ in the mixture at E/p of 100 V cm^{-1} $Torr^{-1}$ is estimated to be around 15 eV by means of the Boltzmann equation analysis.[13] Therefore, the large value of α_m/p at this E/P is attributed to the large σ_i of SF_6 at higher ϵ as seen in Fig. 1. Since σ_i for SF_6 is considerably decreased at lower ϵ, the value of α_m/p decreases with decreasing E/p.[13]

It is concluded from the above discussion that D_{obs} in He mixtures can be interpreted by Eq. (1) and that the discussion on the basis of Eq. (1) is also supported by data on swarm parameters in the mixtures obtained experimentally and theoretically. The self-focusing of the beam is strongly enhanced by the addition of a gas having high IP, small σ_i, and large retardation rate such as N_2. Since O_2 has small σ_i but low IP and slow cooling rate, the effect of O_2 is weak, as seen in Fig. 2.

B. Ar mixtures

Experimental results for Ar mixtures with various additives (1%, 5%, and 10%) have been shown in the previous papers.[3] Only the results for the mixtures containing Xe, N_2, O_2, N_2O, CH_4, C_2H_4, and SF_6 by 10%,
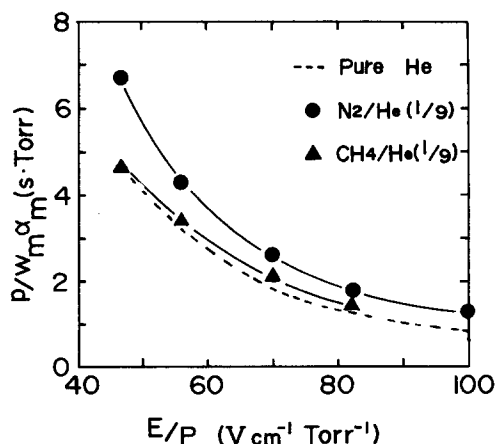

FIG. 3. Values of $p/(w_m \alpha_m)$ estimated by the average electron method for N_2/He (1/9) (●) and CH_4/He (1/9) (▲) as functions of E/p. The value for pure He is shown by ----.
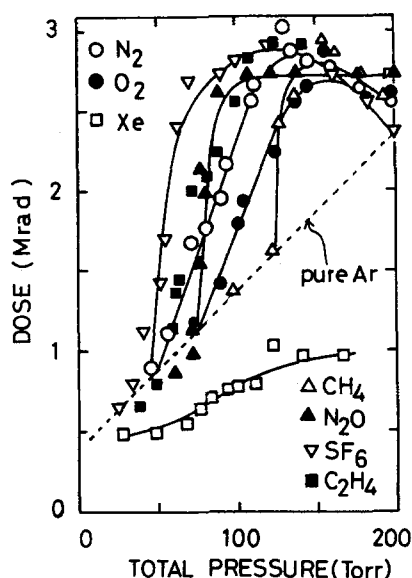
FIG. 4. The dose observed on the beam axis in pure Ar and Ar mixtures as functions of total pressure; --- pure Ar, □ Xe/Ar (1/9), o N_2/Ar (1/9), ● O_2/Ar (1/9), ▲ N_2O/Ar (1/9), ▽ SF_6/Ar (1/9), △ CH_4/Ar (1/9), and ■ C_2H_4/Ar (1/9).

respectively, are shown in Fig. 4. It is noticed that D_{obs} increases sharply from a certain pressure (the on-set pressure) by addition of polyatomic gases in contrast to the case of He mixtures in which the effect of additives appears gradually from 50 Torr and that D_{obs} is decreased by addition of Xe.

As pointed out in the foregoing section, $\bar{\epsilon}$ in Ar is sup-posed not to be reduced from 9 eV down to E/p of a few V cm^{-1} Torr^{-1}. Furthermore, in Fig. 1, Ar has its IP and σ_i which are not as different as He from most of the additives. Therefore, in Eq. (1), the relationship be-tween the first and second terms is not as simple as for He mixtures. The following discussion in considering Eq. (1) is classified as (a) Xe by which D_{obs} is decreased, (b) CH_4 by which D_{obs} is increased only at pressure higher than 125 Torr, (c) O_2 and N_2O by which D_{obs} is increased at pressure higher than about 80 Torr, and (d) N_2, SF_6, and C_2H_4 by which D_{obs} is increased from lower pressure.

1. Xe

The decrease of D_{obs} by the addition of Xe should be explained by the shortened t_i by means of Eq. (1). In Fig. 1, Xe has lower IP and larger σ_i than Ar. Electron energies are not as reduced by Xe as by polyatomic gases for the ns pulse duration. These data support the t_i shortened by Xe added to Ar.

2. CH₄

In Fig. 1, CH_4 has a lower IP and larger σ_i than Ar but not as much as Xe. Furthermore, according to data on D_L/μ,[7] $\bar{\epsilon}$ in CH_4 is supposed as in Ar not to be reduced from 7.4 eV for E/p higher than 20 V cm^{-1} Torr^{-1}. Most of E/p at pressures higher than 50 Torr of Ar lie in this E/p range as seen in Part I.[9] Consequently, $\bar{\epsilon}$ in Ar is not reduced in the presence of the small amount of CH_4 for the major part of the pulse duration, so that t_i is

not lengthened due to the larger σ_i and lower IP of CH_4. However, in increasing the partial pressure of CH_4, some secondary electrons are retarded to an extent lar-ger than in pure Ar due to the larger $Q_t f$ of CH_4 for $\bar{\epsilon}$ above 7.4 eV as seen in Fig. 12 of Part II.[1] Once $F(\epsilon)$ shifts appreciably to the lower-ϵ side so as to affect t_i, this effect on t_i is proportional to the total pressure p in Eq. (1) because α is multiplied by p for α/p given as a function of E/p. Therefore, the curve of D_{obs} rises steeply as seen in Fig. 4 because of the high onset pres-sure. The value of $p/(w_m \alpha_m)$ estimated by the average electron method is almost the same as that for pure Ar at E/p between 80 and 100 V cm^{-1} Torr^{-1}.

3. O₂ and N₂O

Some effect of O_2 added to Ar would be expected be-cause of its small σ_i as is the case of O_2/He for which D_{obs} is affected gradually from lower pressure. How-ever, such an effect is cancelled up to 80 Torr in Ar, which has larger σ_i and lower IP than He. Comparing O_2 with CH_4, σ_i is much smaller for O_2 than for CH_4, IP is the same for both the gases, and $Q_t f$ is not so special for O_2 as for CH_4. These properties are reflected by the difference of D_{obs} for both the gases.

The increase of t_i by addition of O_2 is supported by the Boltzmann equation analysis on the O_2/Ar (1/9) mixture.[15] According to this analysis, the value of pt_i or $p/(w_m \alpha_m)$ increases gradually more than for pure Ar with decreas-ing E/p from 100 V cm^{-1} Torr^{-1} and the increment for the mixture becomes about 10% for E/p around 60 V cm^{-1} Torr^{-1} and, furthermore, 50% for E/p around 30 V cm^{-1} Torr^{-1}.

In Fig. 1, IP for N_2O is almost the same as O_2 and CH_4 and σ_i for N_2O is twice as large as for O_2 at ϵ lower than IP for Ar. Therefore, D_{obs} for N_2O/Ar should be smaller than for O_2/Ar from Eq. (1) if $F(\epsilon)$ is almost the same between both the mixtures. However, in Fig. 4 D_{obs} for the former increases steeply from 75 Torr more than for the latter. This suggests the large shift of $F(\epsilon)$ to the lower-ϵ side and/or the electron attachment pro-cess neglected for Eq. (1). According to Christophorou and Carter,[16] the curve of f for N_2O has the large peak around 2 eV. Therefore, once secondary electrons are slowed down to near thermal by ionizing a N_2O molecule, their acceleration is retarded by N_2O with the above peak of f so as to lengthen t_i in the mixture. On the other hand, the electron-attachment process to N_2O is known at ϵ around 11 eV as well as 2.4 eV.[6] The steeply in-creasing D_{obs} for N_2O/Ar is due to both the effects. The onset pressure of the effect of N_2O in Ar in N_2O/Ar (1/9) is about ten times as large as the pressure at which D_{obs} begins to increase with pressure in pure N_2O.[2]

4. N₂, SF₆, and C₂H₄

In Fig. 1, N_2 has the same IP as Ar and σ_i smaller than Ar. Furthermore, N_2 has the large $Q_t f$ as has been seen in Fig. 13 of Part II.[1] These data suggest that in Eq. (1) t_i is lengthened by addition of N_2 because $F(\epsilon)$ shifts to the lower-ϵ side in the presence of N_2. How-ever, the value of $p/(w_m \alpha_m)$ estimated by the average
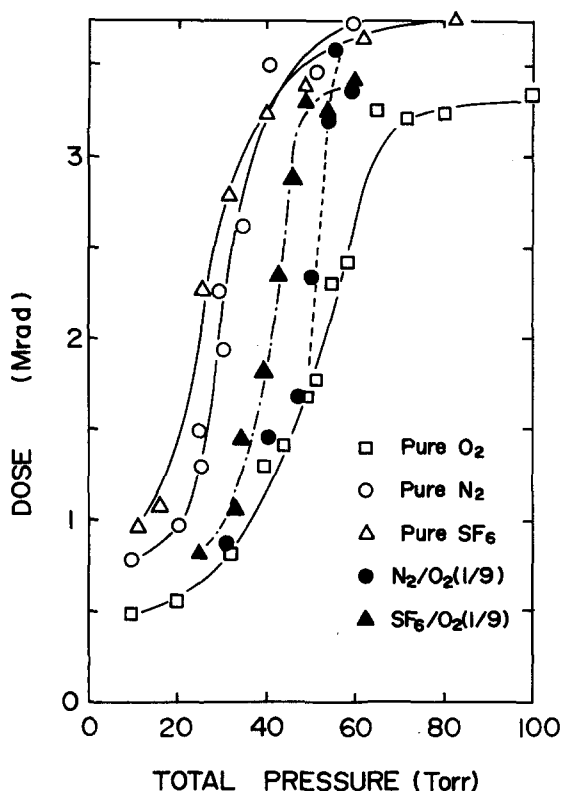
FIG. 5. The dose observed on the beam axis as functions of total pressure for pure O_2 (□), N_2 (○), and SF_6 (△) and also for N_2/O_2 (1/9) (●) and SF_6/O_2 (1/9) (▲).

electron method does not show such a difference between Ar and N_2/Ar.

In Fig. 1, SF_6 has the same IP as N_2 and Ar and the same σ_i as N_2 for ϵ lower than 22 eV while $Q_t f$ is not as large for SF_6 as for N_2 against electrons with energy higher than 5 eV.[1] However, in Fig. 4, the effect of SF_6 appears from pressure rather lower than N_2 and the effect is much stronger than that of N_2. This fact suggests for N_2O that there are the large peak of $Q_t f$ and/or the strong electron attachment process against electrons below 5 eV. According to Kennerly, Bonham, and McMillan,[17] the value of Q_t for SF_6 is 3.6×10^{-15} cm² for 0.5 eV electrons, becomes the minimum (2.2×10^{-15} cm²) for 4 eV ones, and increases gradually up to 3.1×10^{-15} cm² for 12 eV ones. Therefore, the similar retardation effect as in H_2O can be expected against lower-energy electrons. On the other hand, the electron attachment process to SF_6 is known for electrons around 0.4 eV to give efficiently SF_5^-, as well as for thermal. The effect of SF_6 on D_{obs} appears gradually from the lowest pressure (40 Torr) which is lower than the pressure expected from the partial pressure and the pressure from which D_{obs} increases in pure SF_6. Since the phenomena in the present interest are influenced by E/p for which p is not the partial pressure but the total pressure, such an effect from low pressure is due to such a E/p. Since SF_6 has the high IP and the low σ_i relative to N_2O and CCl_2F_2, the effect of the total pressure through E/p must be large for SF_6.

The curve of D_{obs} is almost the same between pure

CH_4 and C_2H_4[11] but quite different between CH_4/Ar and C_2H_4/Ar in Fig. 4. This difference can be interpreted similarly with the He mixtures. According to Yamane,[4] the value of α_m/p for hydrocarbons/Ar at E/p between 5 and 35 V cm⁻¹ Torr⁻¹ increases by addition of a small amount of hydrocarbons, while it decreases with increasing the amount of them owing to decreasing $\bar{\epsilon}$. Although Heylen[18,19] has reported the data for hydrocarbon/Ar, their data contain the Penning ionization processes which should be neglected for the present study.

As seen for N_2O and C_2H_4 in Fig. 4, D_{obs} for most of the mixtures at total pressure lower than the onset pressure was rather smaller than for pure Ar.[3] This may be explained similarly to the case of Xe/Ar by addition of a gas with lower IP and larger σ_i than Ar because ϵ is not reduced sufficiently at its partial pressure.

5. Conclusion

The increase of D_{obs} by addition of polyatomic gases can be explained by the lengthened t_i, which can be estimated qualitatively by Eq. (1). This effect is enhanced by a gas having the higher IP and the smaller σ_i. However, this effect is mainly determined by $F(\epsilon)$. The value of D_{obs} is increased from lower pressure by addition of a gas which retards secondary electrons more strongly. Furthermore, when the electron-attachment process, which is neglected in Eq. (1), occurs for secondary electrons with higher energy, D_{obs} is sharply increased. The discussion on the basis of Eq. (1) is demonstrated for some cases by swarm parameters obtained experimentally or theoretically, but the average electron method was useful only for He mixtures.

C. O_2 mixtures

In Fig. 5, values of D_{obs} for N_2/O_2 (1/9) and SF_6/O_2 (1/9) are plotted by closed marks as functions of total pressure with those for pure O_2, N_2, and SF_6 by open marks. The curve for the mixtures shifts from that of pure O_2 to that of pure N_2 and SF_6; suddenly at 50 Torr for N_2/O_2 and rather mildly from 20 Torr for SF_6/O_2.

As pointed out already, D_{obs} is affected strongly by addition of N_2 to He and Ar. The effect of N_2/O_2 can be explained similarly to that of He and Ar mixtures. However, this effect appears only from 50 Torr, which is the same as the onset pressure for N_2/Ar. This means that t_i is affected appreciably by addition of N_2 higher than 5 Torr due to the shift of $F(\epsilon)$ to the lower-ϵ side.

The difference of the effect between N_2 and SF_6 is already discussed for their Ar mixtures. The difference between both the gases in Fig. 5 should be explained similarly.

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