than about six or seven carbon atoms, the randomisation of hydrogen atoms over the whole chain is unimportant but the elimination of olefins proceeds by complicated reaction steps not yet known. We thus think that, in its present state, the validity of the quasi-equilibrium theory for the prediction of the fragmentation pattern of alkyl radical ions is doubtful.

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80. Effect of Pressure in the Gamma Radiolysis of Cyclohexane in the Vapor Phase

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(3. I. 74)

Summary. The vapor phase γ-radiolysis of cyclohexane has been investigated as a function of pressure, from 100 to 0.5 Torr, in vessels of different dimensions. With decreasing pressure, the yield of hydrogen and substituted cyclohexanes is decreased and the formation of molecules having less than six carbon atoms is enhanced. The curves of yield *vs.* pressure have a sigmoïd shape from which we define a transition pressure depending only on the dimensions of the vessels, the variation of the yield of a given product being the same for all the vessels. We propose and discuss a simple model of competition between the neutralization of the ions in the volume and their diffusion to the surface where they are neutralized. From the slope of the curve of the inverse of the transition pressure *vs.* the square of the characteristic diffusion length, we evaluate a maximum life-time of 68 ms for the ions, using a calculated value of the cyclohexane ions diffusion coefficient.

1. Introduction. – Several authors have explored the γ-radiolysis of cyclohexane vapor at pressures ranging from 170 to 3800 Torr and temperatures from 50° to 300°. *Blachford* and *Dyne* [1], *Theard* [2], *Milhaud* [3] [4], *Sagert & Blair* [5] [6], and *Cosandey* [7] have examined the mechanisms of reactions occuring during the radiolysis of cyclohexane vapor at a fixed pressure by means of different additives and by use of labelled compounds. *Jones* [8] studied this system at pressures and a temperature such that the density was as high as that of the liquid phase (300°, 0.008 to 0.42 g/cm³); with decreasing density the yield of the C_1–C_3 hydrocarbons increased by about one order of magnitude, while the yield of hydrogen and cyclohexene increased only slightly, if at all, and that of bicyclohexyl decreased by a factor of about two. The

proposed explanation for such an effect is either the 'cage' theory of limiting diffusion of the precursors or their collisional deactivation.

Pressure effects in the vapor phase have been observed in other hydrocarbons and are related to the extent of ionic fragmentation, parent-ion deactivation and parent-ion-electron recombination [9]. *Ausloos et al.* [10] studied the pressure effect on the photolysis of cyclohexane at photon energies of 8.4, 10.03, and 11.6–11.8 eV. They extrapolated their results in order to show that in radiolysis (under their experimental conditions) there is no pressure effect on the fragmentation of the parent-ion above 13 Torr. In the present work we have attempted to decrease the pressure to the lowest feasible limit in order to study the radiolysis of cyclohexane at very low pressures.

2. Experimental. – *Materials.* Cyclohexane from two different sources was used:
– *Fluka* puriss., treated with H_2SO_4, NaOH, and $CaCl_2$, and then rectified on a *Podbielnak* column.
– *Phillips* Research Grade from Lot No. 1396, without any further purification. No cyclohexene could be detected by gas chromatography (detection limit: about 10^{-3} mole %). The only impurity found was methylcyclopentane in concentrations of 10^{-3} mole %.

Sample preparation. Cyclohexane lots of about 20 ml, after degassing by several freeze-pump-thaw cycles and drying over 5-A molecular sieve, were stored in a bulb on a vacuum line. Prior to introducing the cyclohexane, the vessels, fitted with a break-seal for product collection, were pumped on the vacuum line and heated with a flame to near the melting point of the Pyrex glass. The metallic cells were pumped overnight while being heated at about 200° in an electric furnace. They were then filled by vacuum distillation to the desired pressure (*Wallace & Tiernan* 50 Torr absolute manometer). The sample was frozen in liquid nitrogen and the vessel then pumped and sealed. Never more than two vessels were filled at the same time in order to avoid a systematic error on the pressure.

Vessels. Seven different types of vessels were used. They are described in table 1, where Λ is the characteristic diffusion length, given by (see e.g. [11]):
$\Lambda^2 = (R/\pi)^2$ for a sphere (A)
$\Lambda^2 = ((\pi/H)^2 + (2.405/R)^2)^{-1}$ for a cylinder.
For the coaxial vessels, a value of the radius R was estimated by the following average:
$R = (R_{in}\theta_1 + R_{ex}\theta_2)/\pi$
where $\theta_1 = \text{Arccos} (2R_{in}/(R_{in} + R_{ex}))$
      $\theta_3 = \text{Arccos} (R_{in}/R_{ex})$; $\theta_2 = \theta_1 + \theta_3$
R_{in} = inner radius of the coaxial vessels (C, E, F)
R_{ex} = outer radius
H   = height of the cylindrical vessels (B, C, D, E, F, G).

Table 1. *Characteristic dimensions of the vessels used* (see text)

Vessel type:	A	B	C	D	E	F	G
R_{in} cm			0.5		1.0	2.0	
R_{ex} cm			2.3		1.95	2.45	
R cm	4.7	2.7	2.0	1.75	1.4	1.1	0.825
H cm		10	15	10	10	10	10
Λ^2 cm^2	2.24	1.13	0.70	0.50	0.34	0.21	0.12
V cm^3	530	245	230	100	82.5	70	25
S cm^2	315	230	290	150	220	345	70
S/V cm^{-1}	0.6	0.94	1.26	1.5	2.7	4.9	2.8

Vessel C is made of two concentric *Kovar* tubes joined by Pyrex glass.

Irradiation. Samples were irradiated with γ-rays from a Gammacell 220 source at a dose rate of about 1 Mrad/h ($6.2 \cdot 10^{19}$ eV/(g·h)). A systematic error, estimated to be 10%, might have been made on the dose rate. All samples were irradiated to 5 Mrad at about 40°.

Analysis. Analyses were carried out on a temperature programmed gas chromatograph equipped with two columns in parallel (Fig. 1). The contents of the vessel were condensed in the sample loop and, after warming up, injected by means of a sample valve. As for the non condensable hydrogen and methane, the quantities injected were aliquots for vessels of the same volume. All the products listed were identified and calibrated by injections of mixtures of known composition. We were unable to find either cyclopropane or propyne. Some minor peaks (such as probably pentane, hexane, dimethylcyclohexane, hexylcyclohexane, etc.), were not identified and not measured.
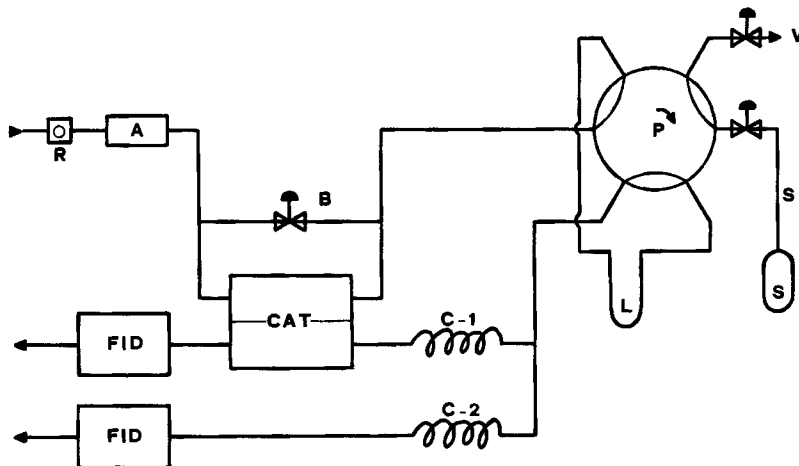

Fig. 1. *Flow diagram of the gas chromatograph*

R: flow controller. – A: 500 ml volume to absorb the injection pressure shock. – C: catharometer detector for H_2 measurement. – B: by pass to balance the flow rate in the catharometer cells. – P: six-port injection valve. – V: to the vacuum line. – S: *Rotulex* joint and connection to breakseal. – L: sample loop with facility to freeze at −196 °C. – C(1): 2,5 m, $1/4''$ out. diameter *Porapak* R column for the lighter products. – C(2): 5 m, $1/4''$ out. diameter *Igepal* CO-880 (nonyl phenoxy polyethylenoxy-ethanol) 15% on Kieselgur for the heavier products.

3. Results. – Fig. 2 shows an example of the pressure effect on the yields, G, of five different products for vessel D. Three distinct pressure regions can be observed: at high pressures, the yield of the products has a constant value which forms a high-pressure plateau. As the pressure is decreased the yield of a given product is either increased or decreased until it reaches another constant value, the low-pressure plateau. The variation of the yield, ΔG, is the same for all the vessels, but occurs at different pressures depending on the geometry of the vessels.

Table 2 gives the average values of the high-pressure plateau yields, G_0, for the seven vessels used, together with the results given by other authors. From the sigmoïd shape of the curves of the yields *vs.* pressure, we define a transition pressure P_{tr}, determined graphically for each product and each vessel, as being the pressure for which $G = G_0 + \Delta G/2$.

For a given vessel we tried to fit an empirical equation relating the G-value of a product to the corresponding pressure in order to linearize the function $G = G(P)$

and obtain values for the transition pressure, $P_{tr,1}$, and for ΔG_1. The equation gives a straight line by plotting $1/(G - G_0)$ *vs.* P^2. This procedure was used for the four vessels B, D, E and F. Table 3 gives the values of the transition pressure for these vessels and for each product, together with the average value of the transition pres-

$$1/(G - G_0) = 1/\Delta G_1 + P^2/\Delta G_1 P_{tr}^2 \tag{1}$$

sure for each vessel. Table 4 gives the average values of G_0, and of the variation of G (determined graphically ΔG, by equation (1) ΔG_1) for each product. From table 4 it can be seen that the graphical and computed values of ΔG are in very good agreement. The values of P_{tr} and $P_{tr,1}$, agree within the limits of their standard errors.
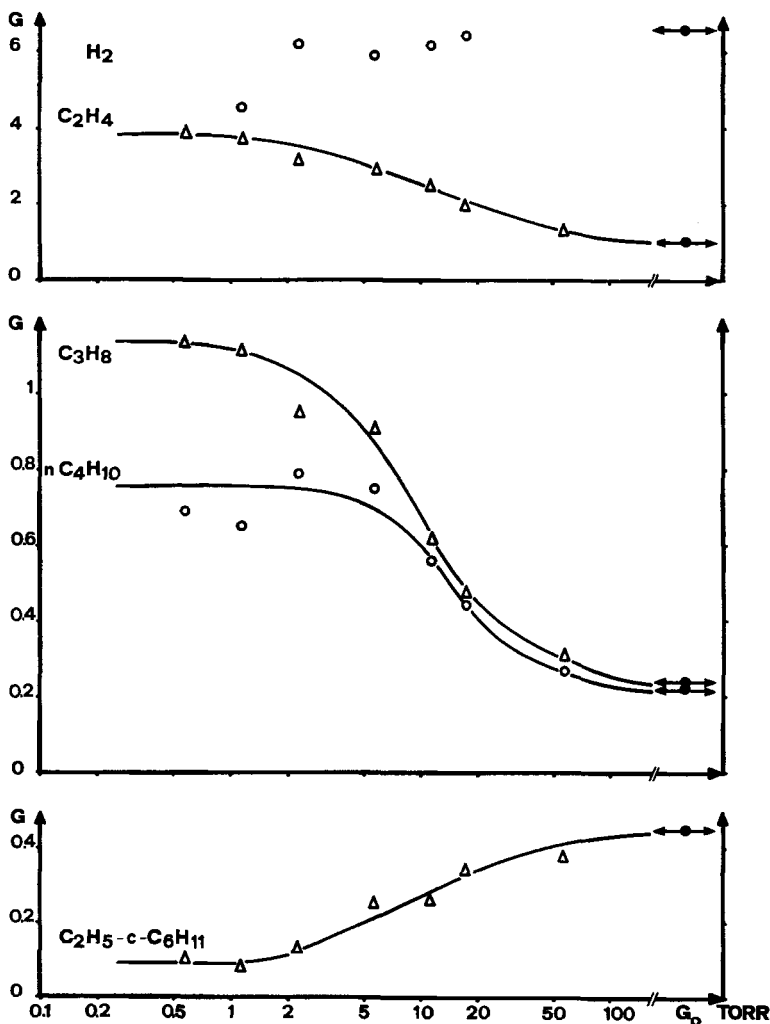

Fig. 2. *Dependence of yield on pressure* (vessel D) G_0 is the high-pressure plateau yield (see text)

The values of P_{tr}, and ΔG for several of the products are not given in tables 3 and 4 although their G_0-value appears in table 2. The values for methane, acetylene, methylcyclohexane, allylcyclohexane and bicyclohexyl were too imprecise with certain vessels.

In the case of three vessels (A, C, D) the low pressure plateau could not be attained for hydrogen because its transition pressure is significantly lower than the mean value for the other products, and, for analytical reasons, we could not use lower pressures. Nevertheless, a value of ΔG of 1.7 can be estimated from the results of the other vessels. The dose effect has been examined by *Milhaud* [4] who found stationary yields for all determined products for doses up to 5 Mrad $(3 \cdot 10^{20}$ eV/g) at 270 Torr,

Table 2. *Values of G_0 obtained by different authors*

	a	b	c	d	e	f
Hydrogen	6.6	3.51	4.2	4.7	5.3	4.3
Methane	0.22		0.20	0.36	0.6	0.4
Ethylene	0.98		0.97	1.35	2.8	
Ethane	0.18		0.19	0.71	1.9	
Acetylene	0.21	0.12	0.18	0.32		
Propylene	0.14	0.70	0.24	0.35	0.43	
Propane	0.24	0.33	0.48	0.30	2.2	
Allene	0.014	0.03	0.01			
i-Butane	0.03		0.25		0.18	
1- + *i*-Butene	0.01	0.17	0.03			
n-Butane	0.22	0.33	0.10		0.04	
trans-Butene-2	0.025	0.16	} 0.04			
cis-Butene-2	0.014	0.12				
Butadiene-1,3	0.007		0.04			
Methylcyclopentane	0.10					
Methylcyclohexane	0.15	0.06				
Cyclohexene	0.50	0.92	0.56	1.0	2.2	2.0
Ethylcyclohexane	0.45	0.23		0.3		
i-Propylcyclohexane	0.09	0.09		} 0.2		
n-Propylcyclohexane	0.054	0.03				
Allylcyclohexane	0.03	0.08				
n-Butylcyclohexane	0.10	0.09				
Bicyclohexyl	0.55	1.03	0.64	0.8	1.0	0.7

a: (this work) $3 \cdot 10^{20}$ eV/g (5.0 Mrad), 40 °C, 'High pressure' plateau
b: [7] $3 \cdot 10^{19}$ eV/g (0.5 Mrad), 70 °C, 250 Torr
c: [4] $0–3 \cdot 10^{20}$ eV/g (0–5 Mrad), 50 °C, 270 Torr
d: [2] $3 \cdot 10^{19}$ eV/g (0.5 Mrad), 100 °C, 1000 Torr
e: [1] $3.6 \cdot 10^{19}$ eV/g (0.6 Mrad), 125 °C, 1100 Torr
f: [5] $2.3 \cdot 10^{19}$ eV/g (0.4 Mrad), 133 °C, 700 Torr

except for cyclohexene for which the yield decreases with increasing dose. We have examined the dose effect in the low pressure plateau at about 2 Torr. The results, reported in table 5, show no dose dependence with the exception of cyclohexene, the yield of which decreases with increasing dose. We therefore used a dose of 5 Mrad and rejected the results of cyclohexene for the pressure dependence.

Table 3. *Transition pressures for different products*

Vessel	A	B	C	D	E	F	G
Ethylene	1.9	5.5	6.5	10	16	19	18
Ethane	2.0	5.0	6.0	8	18	22	10
Propylene	1.8	2.2	5.0	9	13	10	20
Propane	1.7	4.7	6.2	9.5	18	19	32
Allene	1.6	4.0	5.0	9	8	8	13
i-Butane	1.8	3.2	4.4	5	13.5	14	16
1- + *i*-Butene	1.9	2.3	5.0	9	7	9	15
n-Butane	2.4	5.5	5.2	13	19	22	28
trans-Butene-2	2.2	2.2	3.3	9.5	5	5	24
cis-Butene 2	2.1	2.7	3.2	8	6	6	22
Butadiene-1,3	2.1	3.4	4.0	5	9	6	22
Methylcyclopentane	1.7	5.2	4.8	9.5	13.5	11	14
Ethylcyclohexane	1.8	2.1	6.0	8	11	9	19
i-Propylcyclohexane		2.3	7.0	8	9	9	17
n-Propylcyclohexane		2.0	7.0	7.0	8	9	22
n-Butylcyclohexane		4.0	6.5	5	8	9	18
$\overline{P_{tr}}$	1.92	3.5	5.3	8.3	11.4	11.7	19.4
	± 0.06	± 0.3	± 0.3	± 0.5	± 1.1	± 1.4	± 1.4
$\overline{P_{tr,1}}$		4.4		9.4	10.4	14.0	
		± 0.4		± 0.6	± 0.9	± 1.2	

The standard errors on the average values over all vessels (up to 10 measurements/vessel) are given.

$\overline{P_{tr}}$ is determined graphically and $P_{tr,1}$ is computed by equation (1).

Table 4. *Values of G_0 and ΔG for different products*

	G_0		$\overline{\Delta G}$	$\overline{\Delta G_1}$
Hydrogen	6.6	± 0.15	– 1.7	
Ethylene	0.98	± 0.01	2.48 ± 0.15	2.30 ± 0.2
Ethane	0.18	± 0.02	0.52 ± 0.05	0.48 ± 0.05
Propylene	0.14	± 0.02	0.94 ± 0.07	0.94 ± 0.05
Propane	0.24	± 0.02	1.09 ± 0.08	1.07 ± 0.08
Allene	0.014 ± 0.002		0.10 ± 0.01	0.09 ± 0.01
i-Butane	0.030 ± 0.006		0.23 ± 0.02	0.18 ± 0.04
1- + *i*-Butene	0.010 ± 0.004		0.12 ± 0.01	0.10 ± 0.02
n-Butane	0.22 ± 0.05		0.56 ± 0.05	0.62 ± 0.06
trans-Butene-2	0.025 ± 0.008		0.29 ± 0.03	0.32 ± 0.01
cis-Butene-2	0.014 ± 0.004		0.19 ± 0.02	0.19 ± 0.01
Butadiene-1,3	0.007 ± 0.002		0.10 ± 0.01	0.10 ± 0.01
Methylcyclopentane	0.10 ± 0.01		0.28 ± 0.03	0.36 ± 0.05
Ethylcyclohexane	0.45 ± 0.05		– 0.37 ± 0.03	– 0.30 ± 0.02
i-Propylcyclohexane	0.090 ± 0.004		– 0.06 ± 0.01	– 0.05 ± 0.01
n-Propylcyclohexane	0.054 ± 0.002		– 0.03 ± 0.01	– 0.03 ± 0.01
n-Butylcyclohexane	0.100 ± 0.003		– 0.06 ± 0.01	– 0.06 ± 0.01

The standard errors on the average values over all vessels (up to 10 measurements/vessels) are given.

$\overline{\Delta G}$ is determined graphically and $\overline{\Delta G_1}$ is computed by equation (1).

4. Discussion. – In order to interpret our results we would like to propose and discuss a simplified model for the neutralization of the ions produced during radiolysis, and the formation of the resulting stable products, in relation to pressure. We suppose that volume neutralization produces mainly hydrogen and substituted cyclohexanes by radical reactions, while when surface neutralization is involved, we observe an increased formation of molecules having less than six carbon atoms, e. g. ethylene.
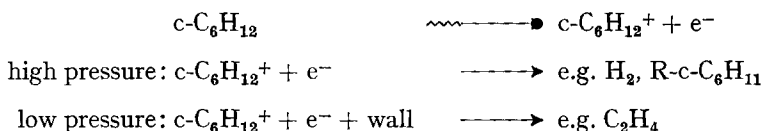
Table 5. *Effects of dose and surface at 2 Torr* (low pressure plateau, vessel D)

Dose in Mrad	0.5	1.0	2.5	5.0	5.0*
Methane	0.38 ± 0.05	0.33	0.30	0.32	0.37
Ethylene	2.9 ± 0.2	2.95	3.30	3.10	3.00
Ethane	0.37 ± 0.1	0.44	0.44	0.52	0.48
Propylene	1.3 ± 0.1	1.2	1.2	1.0	0.9
Propane	0.85 ± 0.1	0.70	0.90	0.95	1.00
i-Butane	0.25 ± 0.05	0.20	0.25	0.26	0.22
1- + *i*-Butene	0.23 ± 0.05	0.25	0.27	0.20	0.14
n-Butane	0.80 ± 0.05	0.65	0.69	0.73	0.60
trans-Butene-2	0.25 ± 0.05	0.25	0.30	0.30	0.32
cis-Butene-2	0.13 ± 0.05	0.12	0.14	0.17	0.18
Methylcyclohexane	0.21 ± 0.05	0.18	0.25	0.29	0.30
Cyclohexene	0.9	0.8	0.8	0.5	0.5
Ethylcyclohexane			0.12	0.13	0.08

* = surface multiplied by a factor of 10.

The standard error is given for 2 to 3 experiments.

The two neutralization paths are in competition with each other, the former being favored at higher pressures, the latter being the only path possible at pressures where the diffusion length is larger than the dimension of the vessel.

$$\text{c-}C_6H_{12} \xrightarrow{\quad\sim\!\!\sim\!\!\sim\!\!\longrightarrow\quad} \text{c-}C_6H_{12}^+ + e^-$$

$$\text{high pressure:}\quad \text{c-}C_6H_{12}^+ + e^- \longrightarrow \text{e.g. } H_2, \text{R-c-}C_6H_{11}$$

$$\text{low pressure:}\quad \text{c-}C_6H_{12}^+ + e^- + \text{wall} \longrightarrow \text{e.g. } C_2H_4$$

The results in tables 3 and 4 show that for a given product the values of ΔG are the same for all the vessels but that the values of the transition pressures depend on the dimension of the vessels. This geometrical dependence shows that, at low pressures, the life-time, τ, of ions is determined by their diffusion to the vessel wall. From low pressure values τ increases with increasing pressure until the concentration of ions and electrons becomes large enough to result in neutralization by volume recombination. Because of this competition, the net lifetime, τ, decreases with still further increase in pressure. In our model the variation of yield is related to the life-time of the ions; we can therefore conclude that this life-time is the same in all the vessels at the transition pressure. Furthermore, the sigmoïd shape of the curves of the yields G *vs.* the pressure (see fig. 2) indicates that the life-time of the ions is roughly maximal at the transition pressure (τ_{max}) because it corresponds to a maximum of the variation of the yield. In typical irradiation conditions τ will depend only on geometry and

pressure (assuming ambipolar diffusion of ions and electrons [12] [13], ionic number density negligible in comparison with the molecule number density and no electric field). In the case where ambipolar diffusion to the wall is the rate determining step (low pressure region), the characteristic diffusion length, Λ, is related to the life-time, τ, of ions and electrons by the ambipolar diffusion coefficient D_a:

$$\Lambda^2 = D_a \tau \tag{2}$$

As the transition pressure obeys a diffusion law, we can, within reasonable error, estimate a maximum life-time, τ_{max}, by the value τ given by the ambipolar diffusion equation at the transition pressure. By introducing pressure into (2) we obtain:

$$\Lambda^2 P/\tau = D_a P = Cst. \tag{3}$$

where $D_a P$ is pressure independent since D_a is inversely proportional to the pressure, and, at the transition pressure:

$$1/P_{tr} = \Lambda^2/(\tau_{max} D_a P) \tag{4}$$

Table 6 gives the values of $1/P_{tr}$ for the different vessels, and figure 3 shows that a plot of $1/P_{tr}$ vs. Λ^2 gives a straight line passing approximately through the origin. By a least mean squares regression of the data in table 6 we obtain $\tau_{max} D_a P = 4.5 \pm 0.3$ cm$^2 \cdot$ Torr. The ambipolar diffusion coefficient can be evaluated for cyclohexane ions with the aid of a combination of the *Einstein* and *Langevin* equations:

$D_a \cong 2D_+ = 2kT\,35.9\,(\alpha\mu)^{-1/2}$, where μ is the reduced mass of the cyclohexane-ion cyclohexane-molecule pair and α is the polarisability of cyclohexane. Taking $\mu = 42.1$ g/mole and $\alpha = 10.9 \cdot 10^{-24}$ cm^3/molecule [14] we obtain:

$D_a P \cong 66$ cm$^2 \cdot$ Torr $\cdot$ s^{-1}, and $\tau_{max} = 68$ ms.

It is now interesting to compare our results with those of a study of *Warman et al.* [13] on the dissociative attachment of thermal electrons by a microwave conductivity technique. They measured the mean life-time of electrons in non-attaching gases (C_3H_8 and CO_2) in order to determine the magnitude of the correction due to diffusion

Table 6. *Dependence of the transition pressure on the characteristic diffusion length*

Vessel type[a]	A	B	C	D	E	F	G
$1/P_{tr}$ Torr^{-1}	0.52	0.29	0.19	0.12	0.088	0.085	0.052
Λ^2 cm^2	2.24	1.13	0.70	0.50	0.34	0.21	0.12

$1/P_{tr} = (0.025 \pm 0.007) + (0.223 \pm 0.007)\,\Lambda^2$ [b])

a)   see table 1.
b)   the errors of the regression line correspond to the standard deviations.

at low pressures. In a cavity having a characteristic diffusion length of 0.71 cm they determined a maximum life-time of 65 ms at a pressure of 10 Torr. At lower pressures, down to 4 Torr, they observed the expected dependence of the diffusion time on pressure, but at higher pressures they attributed the decrease of the life-time to the increasing concentration of ions and electrons, and the resulting ion-electron recombination in the gas-phase.
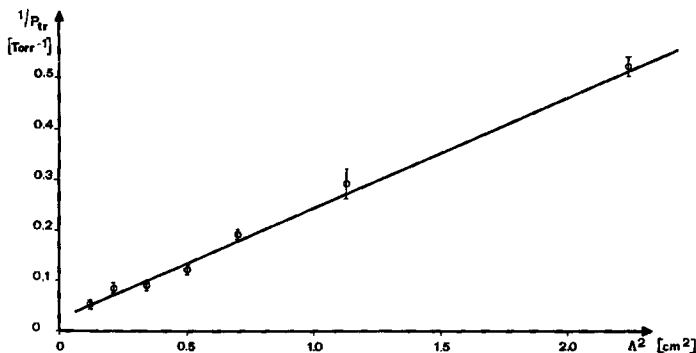
Fig. 3. *Dependence of the inverse of the transition pressure on the square of the characteristic diffusion length*

Woodward & Back found a dose-rate dependence of yield of hydrogen in the γ-radiolysis of different paraffins (C_2H_6, C_3H_8, C_4H_{10}) [15]. They postulated a competition between neutralization in the gas phase at high dose rates (about $2 \cdot 10^{10}$ eV/($cm^3 \cdot s$)) and diffusion of all the ions to the vessel wall at low dose rates (about $300 \cdot 10^{10}$ eV/($cm^3 \cdot s$)). *Klots* [12] applied this simple competition model to *Woodward's* results and found that the rate of diffusion of the positive ions is several orders of magnitude too slow to compete with gas-phase neutralization. He suggests that thermal convection currents enhance the role of surface neutralization.

Experiments with applied electric fields are under way in an attempt to further our understanding of this competition and will be discussed in a subsequent publication.

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