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STUDY OF THE RECOMBINATION IN HEXANE IONIZATION CHAMBERS IRRADIATED WITH GAMMA RAYS

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After a theoretical investigation of the influence of columnar and general recombination on the conductivity of n-hexane irradiated with γ-rays from ^{60}Co, the authors describe the measurements carried out with a view to confirming the theory, with the changing of chamber geometry, of the applied electric field and dose rate. The experimental data are in good agreement with theoretical predictions, at least within the limits of the experiment described.

Finally, a discussion is made of the importance of the results obtained with a view to developing the application in dosimetry of liquid-dielectric ionization chambers.

1. Introduction

In a previous article on ionization chambers filled with n-hexane[1]), we have stressed that the conductivity induced by irradiating the liquid with γ-rays from ^{60}Co increases linearly with the dose rate up to a value of the latter which depends upon the electric field strength between the electrodes. With higher dose rates, a progressive departure from the simple linearity law is observed.

This behavior, which is not predicted in Jaffé's theory on columnar recombination in liquids, was attributed by us to losses by general recombination, which cannot be neglected when the electric field strength is low and the dose rate high. The same effect was also studied by Jahns and Jacobi[2]) by means of a plane ionization chamber, with the changes in the applied electric field strength, in the distance between electrodes and in the dose rate. In interpreting the experimental data, these authors actually take into account the losses due to secondary recombination, as well as the loss caused by primary recombination. To deal with the latter, they use the theory developed by Onsager for dense gases, instead of Jaffé's theory, which they regard as inapplicable.

The results thus obtained are in excellent agreement with the experimental data.

In interpreting the results of our measurements, instead, we preferred to start from Jaffé's theory, in view of the great many experimental confirmations of it, modified to take into consideration also the losses due to general recombination.

2. Theory

Jaffé's theory is based on the assumption that the ions produced by the charged particles passing through fluid, rather than being uniformly distributed throughout the volume involved, are concentrated around the paths of the ionising particles, thus forming cylindrical ion columns, the density of which varies in time, due to diffusion and recombination.

In the case of a plane chamber, calling E the applied electric field strength and φ the angle formed by it with the column considered, the number of ions of the two signs escaping columnar recombination and thus collected in the absence of general recombination will be given by:

$$n' = N_0 d_0 / [1 + \{\alpha N_0 / (8\pi D_0)\} S(x)], \qquad (1)$$

in which N_0 represents the number of ions per unit of length initially present in the column, d_0 the length of the column, α the recombination coefficient, D_0 the diffusion coefficient and $S(x)$ a function which tends to 1 for high values of E, defined by:

$$S(x) = e^x(\tfrac{1}{2}i\pi)H_0^{(1)}(ix),$$

where:

$$x = \tfrac{1}{4}b^2\mu^2E^2(\sin^2\varphi)/D_0^2,$$

being b the initial radius of the column, μ the ion mobility, which is assumed to be equal for positive and negative ions, and $H_0^{(1)}$ Hankel's function of the first species and zero order.

Let now R be the dose rate to which the liquid is exposed. If we assume the irradiation to be uniform throughout the volume occupied by the liquid, the number of columns n_c formed per unit of volume will be:

$$n_c = FR, \qquad (2)$$

where F can be expressed by such parameters as the density of the liquid considered and the energy required to create one pair of ions, and takes into account the unit in which R is expressed.

Under these conditions, the total number of ions per unit of volume which escape columnar recombination

taking into account eqs. (1) and (2), will be given by:

$$n_0 = N_0 d_0 FR/[1+\{\alpha N_0/(8\pi D_0)\}S(x)]. \quad (3)$$

If no general recombination occurs, then the current measured will be:

$$i = n_0 Ze\Omega = N_0 d_0 FRZe\Omega/[1+\{\alpha N_0/(8\pi D_0)\}S(x)], \quad (4)$$

in which Ω is the volume of liquid considered and Ze the charge transported by each ion.

Taking:

$$J(E) = N_0 d_0 FZe/[1+\{\alpha N_0/(8\pi D_0)\}S(x)], \quad (5)$$

which represents the current per unit of volume and of dose rate foreseen in Jaffé's theory, eq. (4) becomes:

$$i = J(E)\cdot\Omega R. \quad (6)$$

Eqs. (5) and (6), originally obtained for α particles, were found to be valid also in the case of radiations not directly ionising, and in particular with γ-rays. Eq. (6), however, as indicated above, is verified in practice only for sufficiently high values of E and low values of R.

To determine a more widely valid expression of eq. (6), let us introduce the assumption that the ions of eq. (3), having escaped columnar recombination, may still undergo general recombination.

In a plane chamber in which are present n_0 ions per unit of volume, taking into account the losses due to general recombination, the collection efficiency will be given by[3]):

$$f = 2/\{1+(1+\xi^2)^{\frac{1}{2}}\}, \quad (7)$$

with:

$$\xi = (\tfrac{2}{3}\alpha/\mu^2)^{\frac{1}{2}}(d/E)n_0^{\frac{1}{2}}, \quad (8)$$

where d is the distance between electrodes, the other symbols have the same meanings specified before, and the mobilities of positive and negative ions continue to be assumed to be equal, for the sake of simplicity.

Then, of the n_0 ions which escaped columnar recombination, only the fraction fn_0 will contribute to the current measured which will then be:

$$i = f n_0 \Omega Ze.$$

Taking into account eq. (5), (7) and (8), we get:

$$i = 2J(E)\Omega R[1+\{1+\tfrac{2}{3}(\alpha/\mu^2)(d^2/E^2)J(E)R/(Ze)\}^{\frac{1}{2}}]^{-1}. \quad (9)$$

Taking:

$$h = \tfrac{2}{3}(\alpha/\mu^2)/Ze, \quad (10)$$

eq. (9) becomes:

$$i = 2J(E)\Omega R[1+\{1+h(d^2/E^2)J(E)R\}^{\frac{1}{2}}]^{-1}. \quad (11)$$

Eq. (11) expresses the looked-for relationship between ionization current and dose rate. For high values of E and low values of R, or when $hd^2J(E)R/E^2 \ll 1$, we find:

$$i = J(E)\cdot\Omega R,$$

as predicted by Jaffé's theory.

We now wish to obtain a relationship similar to

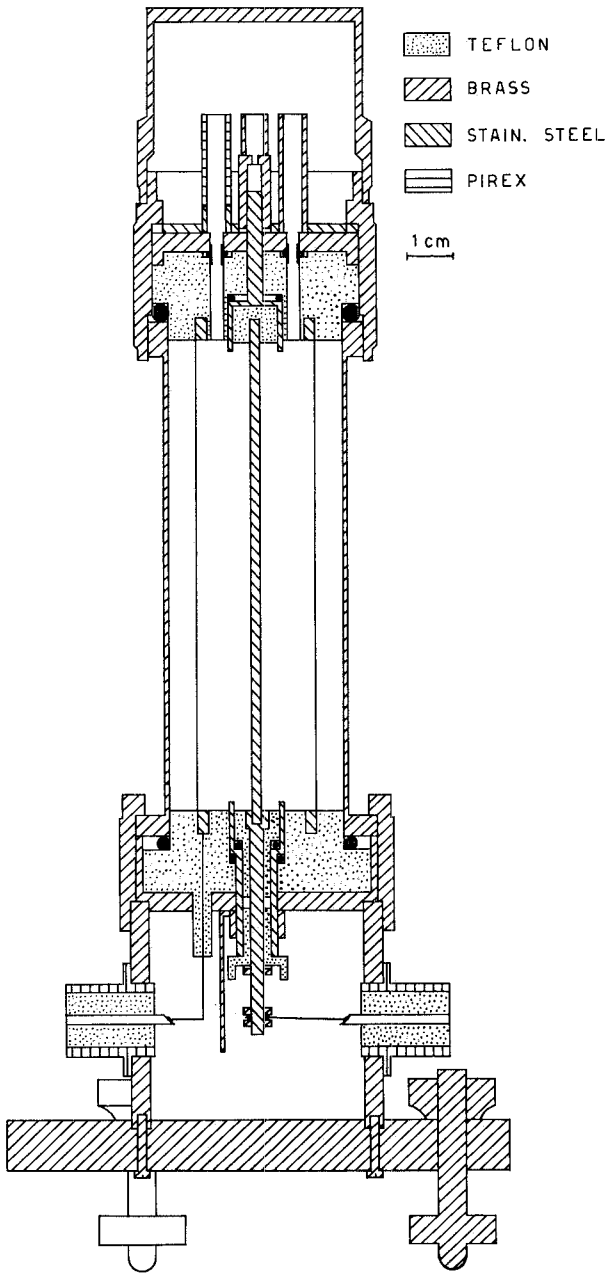

Fig. 1. Cylindrical chamber used in the described experiments. The sensitive volume is about 41.5 cm^3.

eq. (11) also in the case of a cylindrical chamber for, particularly in dosimetry, this geometry is preferred to the plane one.

In this case it will be advisable to bring out the dependences as a function of voltage V rather than of the electric field strength E.

Since Jaffé's theory checked out well also in the case of the cylindrical chamber, in the place of $J(E)$ there will be now a $Y(V)$, which differs from $J(E)$ only in the terms reflecting the different geometry.

In this case, eq. (3) can be written:

$$n_0 = Y(V)R/(Ze) \tag{3a}$$

and eq. (6):

$$i = Y(V)\Omega R. \tag{6a}$$

For the collection efficiency, eq. (7) still holds, where in this case ξ is expressed by:

$$\xi = (\tfrac{2}{3}\alpha/\mu^2)^{\frac{1}{2}}(d_c^2/V)\{Y(V)R/(Ze)\}^{\frac{1}{2}} \tag{8a}$$

and:

$$d_c^2 = (a-b)^2[\{(a/b)+1\}/\{(a/b)-1\}]\cdot\tfrac{1}{2}\ln(a/b),$$

being a and b respectively the radiuses of the external and internal electrodes.

In the place of eq. (9) we then obtain:

$$i = 2Y(V)\Omega R[1+\{1+\tfrac{2}{3}(\alpha/\mu^2)(d_c^4/V^2)Y(V)R/(Ze)\}^{\frac{1}{2}}]^{-1} \tag{9a}$$

and, taking into account eq. (10) we then obtain for current:

$$i = 2Y(V)\Omega R[1+\{1+h(d_c^4/V^2)Y(V)R\}^{\frac{1}{2}}]^{-1}. \tag{11a}$$

Eq. (11a) is entirely similar to eq. (11). In this case, the influence of the general recombination may be neglected only if $hd_c^4Y(V)R/V^2 \ll 1$, and then eq. (6a) will again be obtained, as predicted by Jaffé's theory.

We shall use eqs. (11) and (11a) to interpret our experimental results.

3. Experimental results and discussion

In order to check the validity of the theory outlined above, we have applied eq. (11) to account for the behavior observed when irradiating with γ-rays from ^{60}Co the plane chamber described in a previous paper[4]), and eq. (11a) for the cylindrical chamber, a diagram of which is shown in fig. 1.

With both the plane and the cylindrical chamber we have used as a filling liquid the n-hexane normally used in spectro-analysis, and previously distilled several times to reduce its conductivity. The current as usual, was measured with an electrometer and recorded in graphic form. The dose rate was changed by moving ^{60}Co sources to various distances from the chamber.

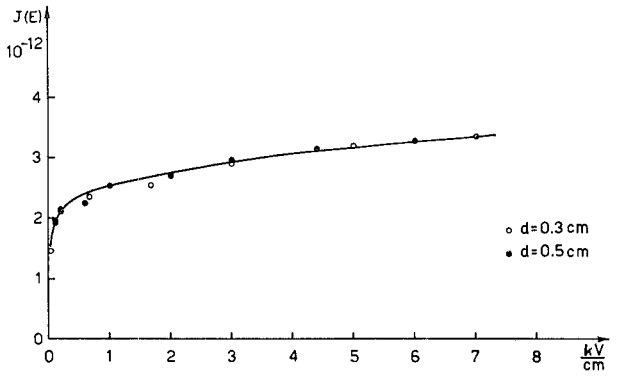

Fig. 2. Function $J(E)$ vs the electric field strength.

In order to compare the experimental results with the theoretical results foreseen in eqs. (11) and (11a), we first determined parameters h, $J(E)$ and $Y(V)$ which appear in these equations.

In order to determine h, we used eq. (10), in which we considered $\mu^2 = \mu_+\mu_-$, being μ_+ and μ_- the mobilities of the positive and negative ions, for which we chose respectively the values of $\mu_+ = 4.5\times10^{-4}$ cm^2/V·sec and $\mu_- = 9\times10^{-4}$ cm^2/V·sec. These are the same values used by Jahns and Jacobi, in good agreement with the experimental values found by Gzowsky[5]). For α we took the value of 1.3×10^{-9} cm^3/sec, deducible from Langevin's theory with the above mentioned mobility values. Finally, assuming $Z = 1$, we obtained $h = 1.34\times10^{16}$ $V^2/A\cdot$cm.

The values of $J(E)$ and $Y(V)$ were instead obtained empirically, in part because of the uncertain knowledge of some of the parameters by which they are defined, but above all due to the lack of experimental data concerning the energy required to create one pair of

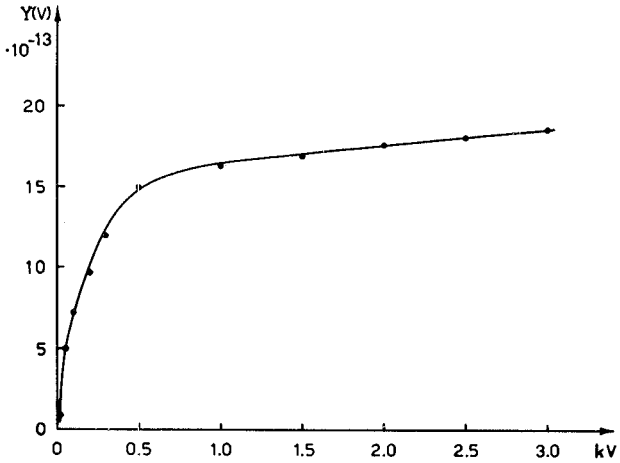

Fig. 3. Function $Y(V)$ vs the collection voltage.

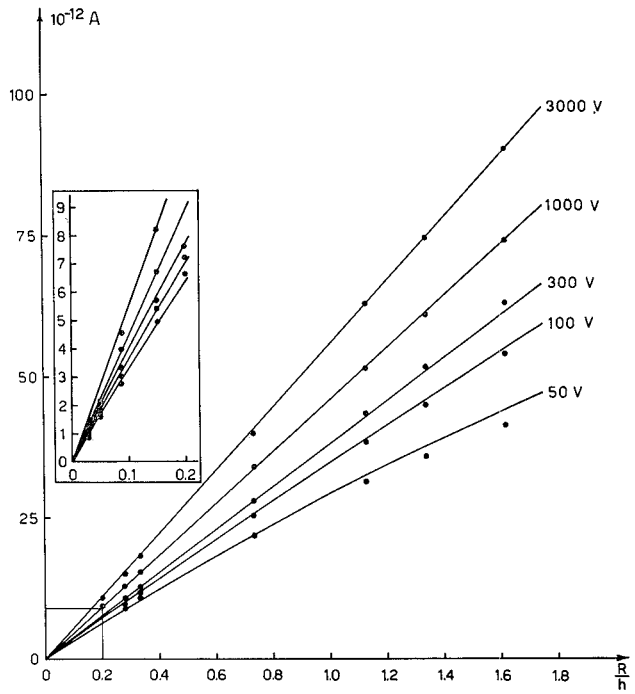

Fig. 4. Theoretical curves and experimental points of the ionization current vs dose rate for some collection voltages in the plane chamber with an interelectrode distance $d = 0.5$ cm. A detail of the figure is shown in the square.

ions in liquid hexane. In general certain authors make up for this lack by using the value relating to hexane in the vapor stage, but we feel that this practice is not entirely free from criticism.

Therefore, to obtain $J(E)$ and $Y(V)$ we carried out ionization-current measurements with low-intensity radiation fields. Under these conditions, the general recombination may be neglected. Jaffé's theory then applies and the values of $J(E)$ and $Y(V)$ can be simply obtained by using eqs. (6) and (6a).

In fig. 2, the values of $J(E)$ thus found, expressed in A/(cm^3 · R/h), are plotted as a function of the applied electric field strength. The measurements were made with two electrode gaps ($d = 0.3$ cm and $d = 0.5$ cm).

Likewise, in fig. 3, relating to the cylindrical chamber, the values of $Y(V)$, expressed in the same unit of measurement, are shown as a function of the applied voltage.

With the h and $J(E)$ values determined as indicated above, we tried to interpret the measurements obtained by us with the plane chamber and presented in a previous paper[1]). The dose range investigated extended from approximately 30 mR/h to approximately 1.62 R/h with $d = 0.5$ cm, and from approximately 50 mR/h to approximately 1.62 R/h with $d = 0.3$ cm.

For dose rates, expressed in R/h, reference was made to measurements in air.

In fig. 4, for $d = 0.5$ cm, the experimental points are compared with the curves predicted by eq. (11). In the dose rate range considered by us, the influence of general recombination is noticeable only with collection voltages under 300 V, while for higher voltages the current always increases linearly with the dose rate.

The theoretical curves and experimental points relating to measurements with $d = 0.3$ cm are shown instead in fig. 5. The response is now linear for voltages above 200 V.

In both cases the agreement between experimental and theoretical results is to be regarded as good, at least within the experimental errors, which are certainly not under 5%.

The measurements with the cylindrical chamber were carried out over a broader dose rate range, extending

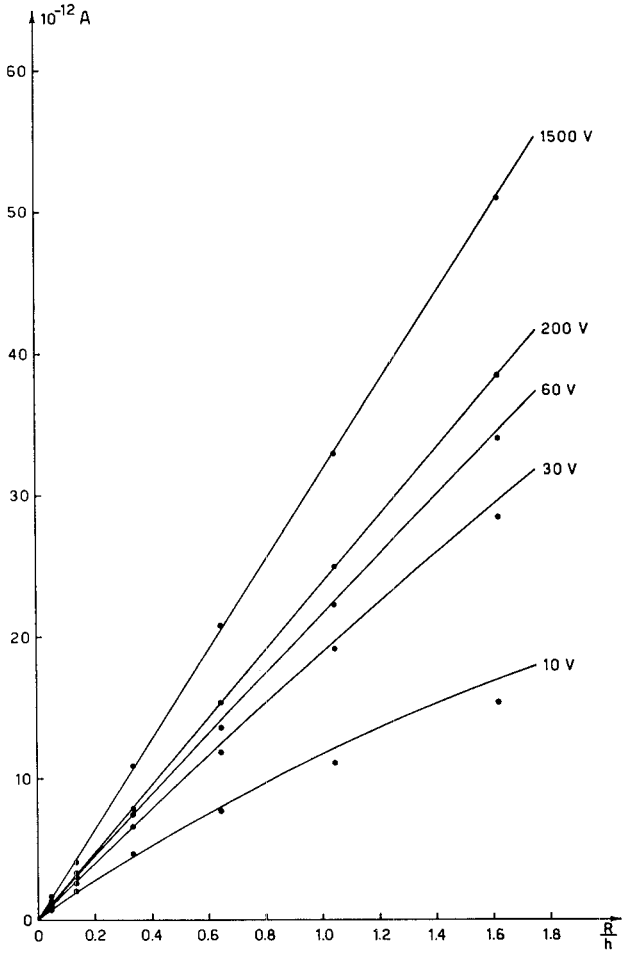

Fig. 5. Theoretical curves and experimental points of the ionization current vs dose rate for some collection voltages in the plane chamber with an interelectrode distance $d = 0.3$ cm.

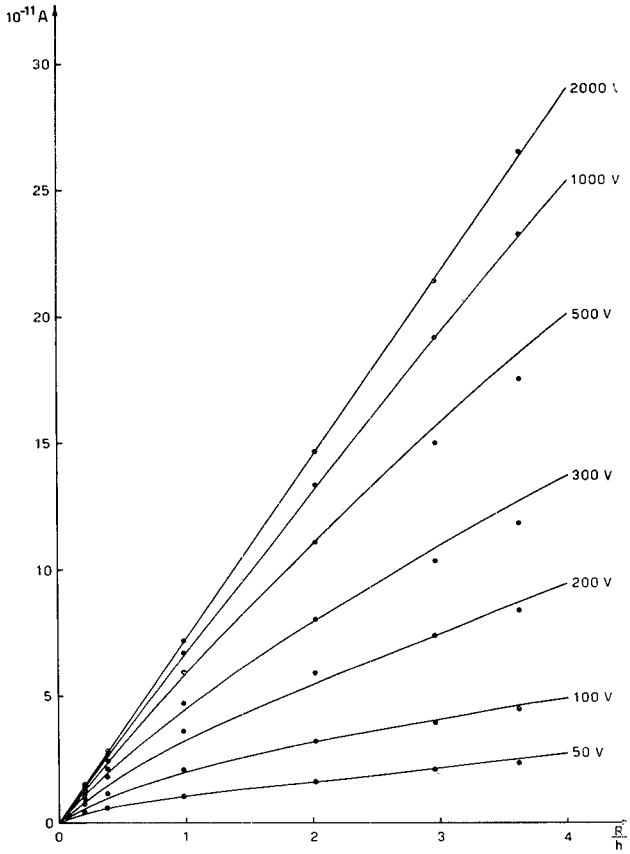

Fig. 6. Theoretical curves and experimental points of the ionization current vs dose rate for some collection voltages in the cylindrical chamber.

from approximately 200 mR/h to approximately 3.62 R/h, with a view to better evidencing the departure from linearity observed as the dose rate increases. The results of the measurements and the theoretical curves predicted by eq. (11a) are shown in fig. 6 for various collection voltages. In this case too the agreement between experimental results seems to be quite satisfactory.

4. Conclusions

As shown in figs. 4, 5 and 6, eq. (11) and eq. (11a) seem to interpret correctly the experimental results, at least within the limits of our experience.

It seems thus proven that the non-linearity observed between ionization currents and dose rate should be attributed to the effect of general recombination which, at high rates and low voltages, cannot be neglected as compared to that of columnar recombination. With sufficiently high electron fields, permitting the collection of all ions escaping columnar recombination, the results predicted by Jaffé's theory are again found.

We wish to point out that the relationships found by us are similar to those established by Jahns and Jacobi for plane chambers; there is the same dose-dependence of the ionization current, while the structure of the electric field strength dependent coefficient appears to be different. This is due to the fact that while we interpreted columnar recombination through Jaffe's theory, those authors used Onsager's theory.

Both processes, however, lead to conclusions that are in satisfactory agreement with the experimental data.

The results of our work, moreover, seem to us to be of particular interest towards the development of dosimetry with liquid-dielectric ionization chambers. In effect, through eq. (11) with a plane chamber and eq. (11a) with a cylindrical chamber, it appears possible to establish the limits of voltage and dose within which we must expect a linear response from a dosimeter based on ionization in a liquid dielectric. Having determined the dose range in which the dosimeter is to operate, the collection voltage will be chosen by requiring condition $hJ(E)d^2R/E^2 \ll 1$ to be met in the case of plane geometry, or condition $hY(V)d_c^4R/V^2 \ll 1$ in the case of cylindrical geometry.

Furthermore, it appears possible to operate also with any suitably low collection voltage, for instance of the same order of magnitude as that used with air chamber, provided in this case recourse is had to eq. (11) or eq. (11a) for the calibration of the instrument.

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