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Initial recombination of ions in ionization chambers filled with hydrocarbon gases

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Abstract. Data on the initial recombination of ions are needed for designing recombination chambers, used in mixed radiation dosimetry in order to obtain information about radiation quality. Nevertheless, experimental data for pressurized gases are scarce, whereas reliable theoretical models are absent. The paper provides experimental data on the initial recombination in hydrocarbon gases (methane, ethane and propane) depending on gas pressure up to 1.8 MPa. The data were derived from the measurements of saturation curves of a tissue equivalent recombination chamber of an REM-2 type, in reference radiation fields of ^{137}Cs and ^{239}Pu-Be sources. Ion recombination due to volume recombination and back diffusion of ions was subtracted from the measured values of ion collection efficiency. The values of gas parameters needed for the subtraction procedure were determined by measurements of saturation curves of the chamber at several dose rates for each gas pressure. The important practical rule resulting from the experiments is that in the first approximation the initial recombination does not depend on the kind of the hydrocarbon gas but only on its density in the chamber.

Key words: initial recombination • ionization chambers

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Introduction

Ion recombination in ionization chambers is usually considered as a side effect, which influences the accuracy of the determination of the absorbed dose. This is generally true for volume recombination, but not always for initial recombination, i.e. the recombination which occurs between the ions created in the same track of a single ionizing particle or resulting from a single ionization event. Contrary to volume recombination, the initial one does not depend on dose rate but it depends on ion density within the tracks. Therefore, the measurable parameters associated with initial recombination carry information about the local density of ions created in the tracks of the ionizing particles in small volumes of the traversed medium (with a mass of the order of one femtogram) and can be correlated with the linear energy transfer (LET) or with other parameters describing radiation quality.

The phenomenon of initial recombination and its relationship with radiation quality has been used in the so-called recombination chambers, where the conditions for the initial recombination are created purposely. The chambers are mostly tissue equivalent, filled with hydrocarbon gases at elevated pressure, in order to keep the gas density in the chamber volume above approximately 2 kg/m^3. The chamber volume, the polarizing voltage and the distance between electrodes in the chamber are optimized in such a way that the ions are

mostly collected on the electrodes before the tracks of ionizing particles diffuse so much that volume recombination can start. Therefore, initial recombination dominates over volume recombination, at least at moderate dose rates. From this point of view, the physical situation is similar to that in liquids and tissues, where volume recombination is practically negligible.

The recombination chambers [15] and several recombination methods [3–5, 7, 13, 14, 16] are now in wide use in radiation protection dosimetry, radiation biology, radiotherapy and in fundamental research. Although the first recombination methods have been described and successfully used many years ago [10, 12, 18], the basic data are still lacking, so as to be able to design recombination chambers with *a priori* given parameters. This is because of complex and usually not calculable dependence of the ion collection efficiency on gas pressure and on electrical field strength in high-pressure ionization chambers.

The aim of this work was to determine the initial recombination in some hydrocarbon gases depending on gas pressure. The gases were selected as being most suitable for filling the tissue-equivalent recombination chambers because the hydrogen content in gas molecules is similar or slightly higher than that in soft human tissue.

Materials and methods

The recombination chamber of REM-2 type with tissue equivalent electrodes [17] was used for the measurements. The chamber has the volume of about 1800 cm^3 and the distance between electrodes $d = 7$ mm. It was consecutively filled with methane, ethane, ethylene and propane up to the maximum pressure of $p_{max} = 1.8$ MPa. The main components of the measuring system are displayed in Fig. 1.

The chamber was connected to an integrated system consisting of a stabilized high voltage supply and a capacitance electrometer, connected to a computer with software for measurement control and data acquisition.

The polarizing voltages, usually from 0.4 V up to 1200 V of both polarities, were applied consecutively to the chamber's electrodes, in order to determine the saturation curve. The procedure was performed at two distances from a ^{137}Cs gamma source and in the field of a ^{239}Pu-Be neutron source (the gamma radiation

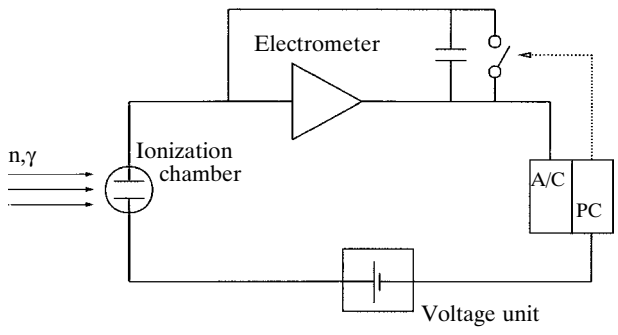

Fig. 1. Block diagram of the measuring system.

contribution to the ambient absorbed dose was about 20% of the total dose). The ionization current at a given polarizing voltage was measured with reproducibility of about 0.1%.

The recombination methods involve determination of the saturation curve, i.e. the dependence of ion collection efficiency on the applied voltage, $f(U) = i(U)/I$, where $i(U)$ is the ionization current measured at the voltage U and I is the saturation current. At a low gas pressure the saturation current can be determined for gamma radiation by commonly used extrapolation methods, based on the theories of initial or volume recombination. The choice of the extrapolation method depends on irradiation conditions. At a gas pressure of above about 1 MPa, extrapolation is difficult and in the case of neutron radiation it becomes practically impossible. Therefore, it is sometimes useful to normalize the measured ionization current values to the ionization current $i(U_S)$, measured at the maximum applied voltage U_S, instead of the more common normalization to the saturation current I. Therefore, two quantities will be used in further considerations – the absolute ion collection efficiency $f(U) = i(U)/I$ and the relative ion collection efficiency $f'(U) = i(U)/i(U_S)$.

The saturation curves were determined for different hydrocarbon gases in dependence on gas pressure, p, radiation dose rate, $\dot{D}$, and gas density ρ. The dependence of initial recombination on the polarizing voltage, U, was derived from the measured saturation curve using the following procedure:

a. The ionization currents measured at opposite polarities of the applied voltage were averaged. Electrical field strength between the electrodes of the chamber always differs from the desired value because some electrical charge is always collected on non-conductive areas of the tissue equivalent electrodes (so-called charge memory effect [15]). There is also a collection of ions generated in parasitic volumes of the chamber (e.g. between the electrode with its connecting part and the chamber housing). The procedure of averaging ionization current values measured for both polarities eliminates these effects, so in this work $i(U)$ always denotes the average value: $i(U) = [i(+U) - i(-U)]/2$.
b. Dark current (measured by the system when the chamber was not exposed to any source of radiation) was subtracted, if its value was not negligible. There are many undesired effects which influence the measurements. Among these are radiation background and leakage current, which are the components of the dark current. The leakage current is caused mainly by the mechanical generation of charges in cables, by the instabilities of applied voltage and by temperature dependent effects. Usually, the leakage current is small and can be neglected. In this work, the dark current had to be subtracted only for the lowest gas density, lowest dose rate and the highest applied voltages, where it constituted almost 1% of the measured current.
c. Parasitic current caused by the back diffusion of ions and by the volume recombination of ions was determined and taken into account. The procedure is described below.

Correction for volume recombination of ions

This work focuses on the determination of the initial recombination of ions, so volume recombination will be considered as an undesired effect. This is why the necessary task was to distinguish between initial and volume recombinations. All the measurements were performed at low dose rates, in order to minimize the volume recombination, but at low voltages volume recombination was unavoidable. In such cases, the ion collection efficiency in the process of volume recombination, f_v, was derived from the measurements of the ionization current at two different dose rates. According to the Boag theory of volume recombination [1], the dependence of f_v on ionization current and polarizing voltage can be expressed by Eq. (1):

$$f_v = \frac{1}{1+\frac{a_v \cdot i}{U^2}} \tag{1}$$

where a_v is a constant for a given chamber and gas filling. It depends on ion mobilities, recombination coefficient and on the distance between the chamber electrodes. The value of this constant can be derived from the measurements at the two different dose rates, $\dot{D}_1$ and $\dot{D}_2$, with the corresponding ionization currents i_1 and i_2, as:

$$a_v = \frac{\Delta f' \cdot U^2}{\left(f_1'(U)\right)^2 \cdot i_1(U_S) - \left(f_2'(U)\right)^2 \cdot i_2(U_S)} \tag{2}$$

where f_1' and f_2' are the relative ion collection efficiencies measured at the dose rates $\dot{D}_1$ and $\dot{D}_2$, respectively, and $\Delta f' = f_2' - f_1'$. Equation (2) can be derived directly from Eq. (1), taking into account the definition of f' (given in the previous section) and assuming that there is no volume recombination at highest voltage, U_S, applied to the chamber.

The values of the factor a_v were determined using Eq. (2) for several polarizing voltages at each gas pressure and then averaged over the voltage region where $0.01 < \Delta f' < 0.1$, corresponding to the highest accuracy. An example is given in Fig. 2 for the chamber filled with methane under the pressure of 690 kPa (with the gas density of about 4.6 kg/m^3), irradiated in the field of ^{137}Cs gamma radiation at two distances from the source – 100 and 300 cm with an ambient absorbed dose rate of 5.2 and 0.57 mGy/h, respectively.

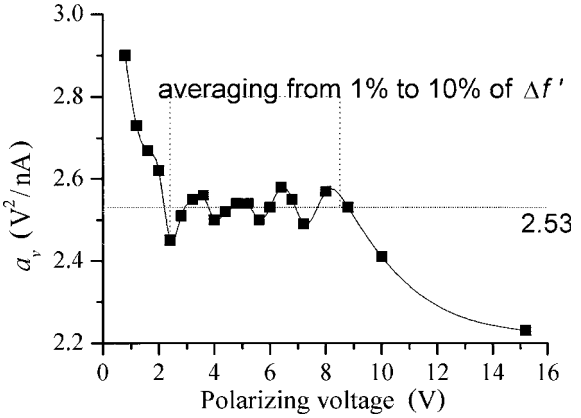

Fig. 2. Averaging the coefficient of volume recombination correction a_v (REM-2 chamber filled with methane, p = 690 kPa).

For high voltages, the correction for volume recombination could be neglected. For $U > \sim 10$ V, volume recombination affected less than 1% of the total electrical charge generated in the chamber at the highest dose rate used, but for voltages of a few volts it reached more than 10% of the generated charge.

Correction for back diffusion of ions

Ions created in the recombination chamber mostly move according to the direction of the electrical field, but some small part of ions migrates also in the opposite direction. This effect, called back diffusion of ions, can be described by relationship (3), introduced by Langevin [9] and later confirmed for parallel-plate air-filled ionization chambers by Böhm [2] and Takata [11].

$$f_d = 1 - \frac{2kT}{eU} + \frac{2}{\exp(eU/kT) - 1} \tag{3}$$

where: f_d is the ion collection efficiency for the process of back diffusion of ions: e is the electron charge: k is the Boltzmann constant and T is the absolute temperature.

For voltages above 0.2 V and for the temperature around 18°C, this relation could be reduced to a simple relation (4):

$$f_d = 1 - \frac{50\ \text{mV}}{U} \tag{4}$$

The measured ion collection efficiencies were corrected for volume recombination and back diffusion of ions using Eq. (5), in order to determine the ion collection efficiency due to initial recombination only, f_{ini}':

$$f_{ini}'(U) = f_2'(U) \cdot \left(1 + \frac{a_v \cdot i_2(U)}{U^2}\right) \cdot f_d \tag{5}$$

An example of the results of the correction for volume recombination and for the back diffusion of ions is shown in Fig. 3, where the upper curve represents the ion collection efficiency resulting from initial recombination only. As can be seen in Fig. 3, no corrections are needed for back diffusion and for volume recombination at dose rates used in this work for voltages above about 20 V.

Results

A full set of the results is presented in Fig. 4 in the form of saturation curves for different gases and two radiation fields (gamma radiation of a ^{137}Cs source and neutron + gamma radiation of a ^{239}Pu-Be source). The

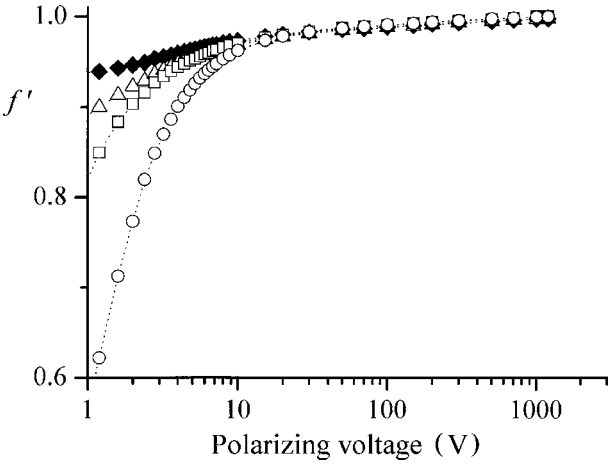

Fig. 3. Relative ion collection efficiency measured by the REM-2 chamber filled with methane (p = 690 kPa) and irradiated at two dose rates $\dot{D}_1$ = 5.2 mGy/h (–○–) and $\dot{D}_2$ = 0.57 mGy/h (–□–). Two upper curves represent the results after correction for volume recombination (–△–) and also for back diffusion of ions (the upper curve).

saturation current for gamma radiation was obtained by the commonly used linear extrapolation of the $1/i$ vs. $1/U$ plot. The results for neutrons are presented as relative ion collection efficiency, i.e. the ratio of the measured ionization current to the current determined at a maximum applied voltage, because at high pressures all the measured values were far from saturation and the extrapolation to the saturation current would introduce excessively large uncertainty.

All the measurements were performed at two or more dose rates in order to investigate the dependence of volume recombination on gas pressure. The results showed that the values of the coefficient a_v of Eq. (1) are a linear function of gas density above 2 kg/m^3 for all the gases investigated (Fig. 5). This relationship can be explained considering the theoretical relation between a_v and parameters of gas molecules [1] and taking into account data showing the linear dependence of the recombination coefficient on the gas density at high pressures [8]. The simple dependence of a_v on the gas density (hence on the chamber sensitivity) makes it possible to determine the a_v value only ones, and recalculate it according to actual sensitivity of the chamber.

Recombination chambers are designed for dosimetric measurements in mixed radiation fields and it is preferable if they have similar sensitivity to gamma and neutron radiation, i.e. the relative neutron sensitivity, h_n, should not differ much from unity. The following definition is used:

$$h_n = A_n/A_C \tag{6}$$

where: $A_n = i_n(U_S)/\dot{D}_n$ is the sensitivity of the chamber to neutrons with spectrum considered; $\dot{D} \equiv D^*(10)$ is the ambient absorbed dose rate at the point of measurement; $A_C = i_C(U_S)/\dot{D}_C$ is the sensitivity to the reference gamma radiation (here: the gamma radiation from a ^{137}Cs source).

The quantity h_n is needed when the chamber is used for the determination of the dose components in mixed

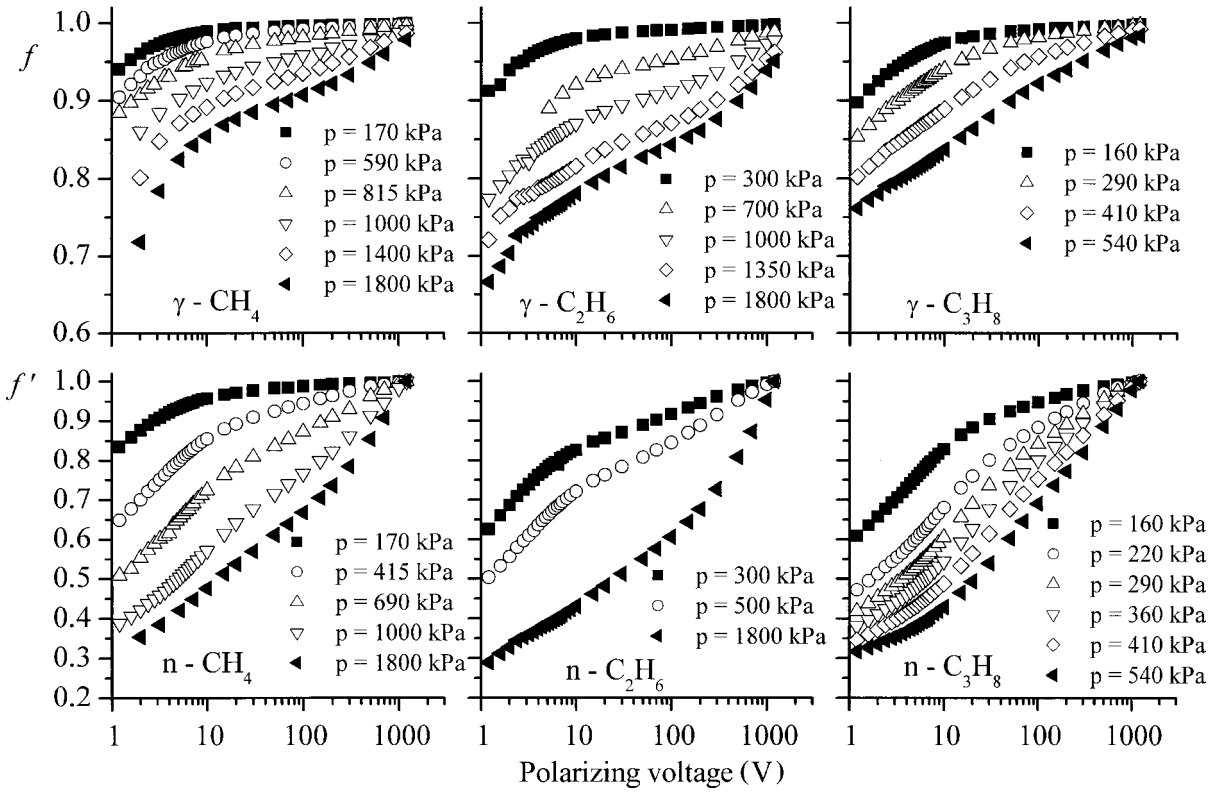

Fig. 4. Ion collection efficiency for gamma radiation of ^{137}Cs source (upper plots) and the relative ion collection efficiency for neutron radiation of ^{239}Pu-Be source (lower plots) determined by the REM-2 ionization chamber filled with methane, ethane and propane. All curves concern initial recombination, so they do not depend on dose rate.

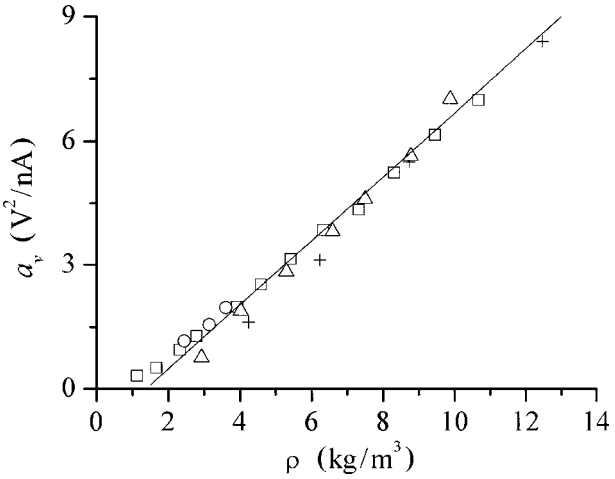

Fig. 5. Dependence of the coefficient a_v on the gas density (squares for methane, crosses for ethane, circles for ethylene and triangles for propane).

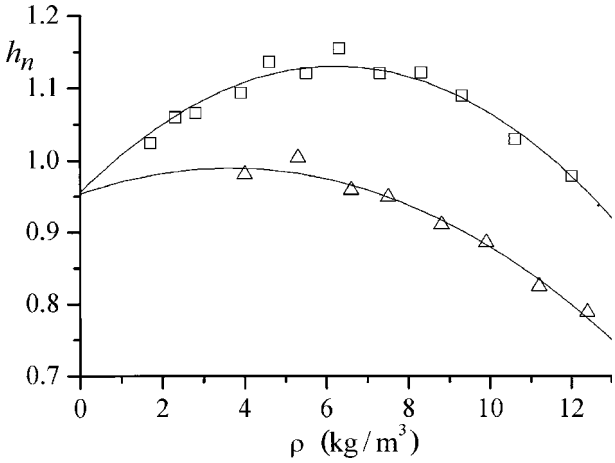

Fig. 6. Dependence of relative neutron sensitivity (neutrons from the ^{238}Pu-Be source) of the REM-2 chamber on filling gas density for methane (the upper curve) and propane (the lower curve).

(neutron + gamma) radiation field, using the twin-chambers techniques. The second chamber is usually a hydrogen-free chamber with low relative sensitivity to neutrons. The dependence of h_n on gas density was determined according to relation (6), using reference radiation fields of ^{137}Cs and ^{239}Pu-Be sources and subtracting the earlier known contribution of gamma radiation from the Pu-Be source, thus h_n concerns neutrons only. The results are shown in Fig. 6.

The gases with a high content of hydrogen (e.g. CH_4) are most convenient in the designing of high pressure recombination chambers used in mixed radiation fields, because with such gases it is more easily to obtain a similar sensitivity of the chamber to neutron and gamma radiations. However, it is not possible to have $h_n \approx 1$ for all neutron energies. For the REM-2 chamber filled with methane (p = 1 MPa, U_S = 1200 V) the value of h_n changes from 0.6 up to 1.5 in the neutron energy range from thermal to 200 MeV [6]. Sometimes, e.g. in the case of unknown neutron energy, it is preferable to have h_n less energy dependent, even if the value of h_n considerably differs from unity.

Conclusions

The results shown in Fig. 4 provide basic data, which can be directly used in designing special recombination chambers, according to the needs specified by the user. The important practical rule is that in the first approximation, the shape of the saturation curve does not depend on the kind of the hydrocarbon gas but only on its density in the chamber. There is also no need to use gases of high purity. The experiments showed that the addition of a few percent of air (by weight) to hydrocarbon gases caused only very small changes in the shape of the saturation curves.

For the measurements at low dose rates e.g. at workplaces and in the environment, the chambers should have high sensitivity but the required accuracy of the measurements is relatively low. Therefore, the higher gas density, above 12 kg/m^3 can be used; however it is practically impossible to obtain the same sensitivity for gamma radiation and for fast neutrons at such gas densities.

At gas densities of above 2 kg/m^3, the coefficient a_v, used in calculations of the correction for volume recombination, is approximately proportional to the gas pressure.

The chambers for LET spectrometry should be filled with a tissue equivalent gas or with a quasi-tissue-equivalent gas (e.g. C_2H_6 or C_3H_8), up to the pressure corresponding to the gas density of 4–8 kg/m^3.

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