Free ion yield observed in liquid isooctane irradiated by $\gamma$ rays. Comparison with the Onsager theory

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Abstract

We have analyzed data on the free ion yield observed in liquid isooctane irradiated by $^{60}\text{Co}$ $\gamma$ rays within the framework of the Onsager theory about initial recombination. Several distribution functions describing the electron thermalization distance have been used and compared with the experimental results: a delta function, a Gaussian type function and an exponential function.

A linear dependence between free ion yield and external electric field has been found at low electric field values ($E < 1.2 \text{ MV/m}$) in excellent agreement with the Onsager theory. At higher electric field values we obtain a solution in power series of the external field using the Onsager theory.

Key words: free ion yield, Onsager theory, isooctane, liquid-filled ionization chamber
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1 Introduction

Liquid filled ionization chambers are currently used in radiotherapy both for dosimetry ([1], [2], [3], [4]) and portal imaging [5]. One of the most commonly used liquids is isooctane (2,2,4 trimethylpentane). This nonpolar liquid has a quite constant stopping power ratio to water in a very wide energy spectrum (less than 3% variation from 0.1 MeV to 20 MeV [1]) and also its intrinsic mass density allows to achieve a spatial resolution in the millimeter range for therapy beams [5].

Free ion yield $G_{fi}(E, T)$, is defined as the number of electron-ion pairs escaping initial recombination per 100 eV of absorbed energy, and experimentally it is obtained from ionization current measurement. The knowledge of how it varies with temperature $T$, with external electric field $E$, and with radiation type, constitutes a fundamental problem to understand the operation of these devices. These dependences have been measured in a large number of liquids ([6], [7]). The Onsager theory [8] describes $G_{fi}(E, T)$, and has been tested in several liquids with good results (see for example [7]). The Onsager theory predicts a linear relationship between ionization current and electric field at low electric field values. The previous dependence can be obtained from numerical resolution of the Onsager theory. This linear behavior has to be extrapolated to very low electric field strength region because volume recombination depletes free charge density produced by radiation in the liquid.

In the current work we describe a detailed method to apply and to test the Onsager theory at the low field strength region and to obtain a precise dependence of $G_{fi}$ with electric field in the linear region for liquid isooctane.

2 Theoretical considerations

2.1 Onsager theory of initial recombination

When ionizing radiation interacts with a liquid, electrons released from molecules thermalize at a distance $r$, where electron and positive ion are still bounded by the Coulomb interaction. This will cause the recombination of the primary ionization pairs produced, which is called initial recombination. These effects are much more relevant in liquids than in gases due to the fact that mass density of liquid hydrocarbons is almost three orders of magnitude higher than density of gases at normal conditions.

Onsager solved the problem of the Brownian movement of an electron under
the influence of both the ion Coulomb attraction and an external electric field $E$ [8]. For isolated ionizations, initial recombination escape probability of an electron-ion pair within the Onsager theory is

$$\Phi(r, E, \Theta, T) = \exp\{-\frac{E_r}{E_0 r_0}(1 + \cos \Theta)\}$$

$$\times \int_{r_0/r}^{\infty} J_0 \left[ 2 \left\{ -\frac{E_r}{E_0 r_0}(1 + \cos \Theta) s \right\}^{1/2} \right] ds$$

where $r$ is the initial separation between electron and ion (i.e. the thermalization distance), $\Theta$ is the angle between the line that initially connects the electron-ion pair and the external electric field. The variables $r_0 = e^2/4\pi\epsilon\kappa T$ and $E_0 = 2\kappa T/e r_0$ are the Onsager radius (the distance at which Coulomb energy equals thermal energy $\kappa T$) and the Onsager field (the field that would produce a voltage $2\kappa T/e$ over a distance $r_0$). Here $\epsilon$ is the liquid dielectric constant ($\epsilon = 1.94 \cdot \epsilon_0$ for liquid isoctane at room temperature), $T$ is its temperature and $\kappa$ is the Boltzmann constant. Finally, $J_0$ denotes the zeroth-order Bessel function.

Mozumder [9] converted the integral of equation (1) in an infinite series using properties of the Bessel functions. He also eliminated the angular dependence averaging over a uniform distribution of $\cos \Theta$. Then, the angle averaged escape probability takes the following form,

$$\Pi(r, E, T) = 1 - \frac{E_0 r_0}{2 E_r} \sum_{n=0}^{\infty} A_n \left( \frac{2 E_r}{E_0 r_0} \right) A_n \left( \frac{r_0}{r} \right)$$

where $A_n(x)$ is the $n$ order incomplete $gamma$ function, which is given by:

$$A_n(x) = \exp(-x) \sum_{k=2n+1}^{\infty} \frac{x^{k-n}}{(k-n)!} = \exp(-x) \sum_{m=n+1}^{\infty} \frac{x^m}{m!}$$

$$= 1 - \exp(-x) \left[ 1 + x + \frac{x^2}{2!} + \cdots + \frac{x^n}{n!} \right]$$

The next expression is more practical for numerical computation of $\Pi(r, E, T)$:

$$A_{n+1}(x) - A_n(x) = -(x^{n+1}/(n + 1)!) \exp(-x)$$

$$A_0(x) = 1 - \exp(-x)$$
Equation (2) is the most adequate formula for calculating escape probabilities for arbitrary values of initial separation and external electric field. In fact, it will be the formula that we will use for numerical calculations of escape probabilities. The expansion in power series of the external field is implicit in equation 2. If we operate in equation (2) then we can obtain it explicitly:

\[
\Pi(r, E, T) = \exp(-r_0/r) \left[ 1 + \sum_{n=1}^{\infty} \left( \frac{E}{E_0} \right)^n B_n(r/r_0) \right]
\]  

(6)

where \(B_n(x)\) is a polynomial of order \(n - 1\) in \(x\), which takes the following form:

\[
B_n(x) = \sum_{m=1}^{n} \left[ \left( \sum_{k=m}^{n} F^n_k \right) \frac{x^{(n-m)}}{m!} \right]
\]  

(7)

The numerical coefficients \(F^n_k\) were calculated by Mozumder [9], and they are given by

\[
F^n_k = 0 \quad \text{for } k > n
\]  

(8)

\[
F^n_n = \frac{2^n}{(n + 1)!}
\]  

(9)

and

\[
F^n_{k-1} = F^n_k + \frac{(-1)^{n-k+1}2^n}{k!(n-k+1)!} \quad \text{for } k \leq n
\]  

(10)

We must keep in mind that thermalization distance is not the same for all electrons. Due to this fact a distribution function \(f(r)\), such as to \(\int_0^{\infty} f(r)dr = 1\), is usually introduced ad hoc to describe electron thermalization distances. Several distribution functions had been used by different authors in the literature (see for example [7] and [9]). In the current article we will test the three most used:

- delta function

\[
f(r, \rho) = \delta(r - \rho)
\]  

(11)

- Gaussian type function

\[
f(r, \rho, \sigma) = \begin{cases} 
N \exp[-(r - \rho)^2/\sigma^2] & \text{if } r \geq 0 \\
0 & \text{if } r < 0
\end{cases}
\]  

(12)
• exponential function

\[ f(r, \rho) = \begin{cases} \frac{1}{\rho} \exp(-r/\rho) & \text{if } r \geq 0 \\ 0 & \text{if } r < 0 \end{cases} \] (13)

The first and second distributions are characterized by a single parameter \( \rho \). The Gaussian type distribution, in addition, has a dispersion parameter \( \sigma \). However, in order to obtain a single parameter distribution we will correlate both parameters. In advance we will take \( \sigma = 0.25 \cdot \rho \) such as [9]. This choice provokes that the difference between the normalization factor \( N \), and the normal Gaussian normalization factor \( 1/\sqrt{2\pi\sigma} \), be less than 0.1%.

Within this framework we can write the escape probability averaged over the thermalization distances as:

\[ P_{\text{esc}}(E, T) = \int_0^\infty \Pi(r, E, T) f(r) \, dr \] (14)

An interesting property of the Onsager series is that the first expansion term of the escape probability does not depend on the thermalization distance \( r \). From equations (6) and (7) we can derive that when the electric field is sufficiently low to verify

\[ 3\left(\frac{E_0}{E}\right) \gg 1 - 2\left(\frac{r}{r_0}\right) \]

the \( n = 1 \) term is much higher than the \( n = 2 \) term in equation (6) (and hence than terms \( n > 2 \)). Then we can truncate the series to first order and so the escape probability rises linearly with the electric field:

\[ \Pi(E, T) = \exp(-r_0/r) \left(1 + \frac{E}{E_0} + \cdots\right) \] (15)

In this case the intercept-to-slope ratio \( E_c \) is predicted to be the Onsager field \( E_0 \):

\[ E_c = E_0 = \frac{8\pi(\kappa T)^2}{e^3} \] (16)

This result is a powerful test to compare experimental data with the Onsager theory because it does not depend on the distribution function used to describe the electron thermalization distance.
Fig. 1. Escape probability calculated using a Gaussian distribution (continuous line), an exponential distribution (dotted line) and a delta distribution (dashed line), plotted against electric field for liquid isooctane at $T = 294$ K.

As we will see later, for isooctane at room temperature irradiated by $\gamma$ photons, $r/r_0 \sim 0.6$ and the linear approximation (15) is right at low electric fields. Figure 1 shows escape probability variation with external electric field for the case of liquid isooctane at $T = 294$ K. Figure 2 shows the low electric field region where a linear relationship is expected.
2.2 Free ion yield calculation within the Onsager framework

Free ion yield, $G_{fi}$, is defined as the number of electron-ion pairs created (i.e. those escaping initial recombination) in the ionization medium per 100 eV of absorbed energy. This magnitude plays a similar role to the $W$ factor in gases. However, $W$ is constant and free ion yield depends on temperature, on external electric field and on radiation type as escape probability does. Within the Onsager theory we can write

$$G_{fi}(E, T) = N_{tot}P_{esc}(E, T)$$

(17)

where $N_{tot}$ is the total number of electron-ion pairs formed initially in the ionization medium (i.e. before initial recombination) per 100 eV of absorbed energy, and $P_{esc}$ is the escape probability described in the previous subsection.

Free ion yield at zero external electric field is denoted as $G_{fi}^0$, and takes the following form:

$$G_{fi}^0 = N_{tot} \int_0^\infty f(r) \exp(-r_0/r)$$

(18)

If we are working at electric field values for which the relation between escape probability and electric field is linear, then we can write

$$G_{fi} = G_{fi}^0 + aE$$

(19)

and the intercept-to-slope ratio from equation (16)

$$E_c = \frac{G_{fi}^0}{a} = E_0$$

(20)

3 Experimental results

In order to measure the ionization current from an isooctane layer under irradiation we built a square shaped parallel plate liquid ionization chamber. The chamber walls were fabricated using FR4 fiber glass reinforced epoxy copper clad on both sides, covering a total area of 4.08 cm × 4.72 cm. The FR4 thickness was 0.8 mm while the copper layer was 35 µm thick. The two chamber walls were glued on both sides of an epoxy plate spacer to provide the 0.8 mm isooctane gap. To guarantee a constant dose rate along the gap, the detector
was inserted between two PMMA$^2$ plates of 5 mm and 5.8 mm thickness, the first one on the incident beam side. As ionization medium we used Merk’s liquid isooctane$^3$ with an estimated purity of 99.8%. Figure 3 shows a scheme of the device cross section.

Experimental tests of the chamber were made in the $^{60}$Co unit of the Complexo Hospitalario Universitario de Santiago (CHUS). The radiation field was set to 10 cm×10 cm at the isocenter$^4$ to cover the whole chamber. In order to obtain isooctane free ion yield we measured the ionization current produced in the whole chamber using a nanoammeter Phillips Fluke PM 2525, for several polarization voltages. High voltage was supplied by a CAEN N471A NIM module.

Distance between the detector and the cobalt source was set to 130 cm (equivalent to a dose rate around 0.4 Gy/min). The dose rate was chosen to have a negligible volume recombination in the upper part of the ionization current vs. voltage curve. Figure 4 shows the experimental data obtained.

If volume recombination and space charge effects can be ignored, the ionization current is proportional to the number of electron-ion pairs released in the liquid per unit time and unit volume $N_{in}$ (initial recombination is included in $N_{in}$):

$$I = e h A N_{in}$$

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$^2$ Polymethylmetacrylate.
$^3$ Isooctane Merk Uvasol quality grade.
$^4$ 80 cm from the cobalt source.
where $e$ is the electron charge, $h$ is the isooctane gap and $A$ is the detector area. From this equation the free ion yield can be calculated as

$$G_{fi} = \frac{I}{e \Delta \varepsilon} \quad (22)$$

where $\Delta \varepsilon$ is the energy deposited in the medium per second. In this case $\Delta \varepsilon = (4.79 \pm 0.19) \cdot 10^{11} \text{ 100 eV/s}$, that was calculated through the EGSnrc code.

To apply equation (22) we require a charge collection efficiency higher than 99% and also that field screening be negligible. To calculate this limit we used a numerical simulation of the charge carriers transport. When the distance between the cobalt unit and the detector is 130 cm, and the polarization voltage is higher than 600 V, general charge collection efficiency is higher than 99%. This agrees with the Greening theory ([10], [11]) about general charge collection efficiency. Within this theory the polarization voltage that must be applied to obtain an efficiency $f$, is

$$V^2 = \frac{1}{6} \frac{m^2 h^4 e N_{in}}{(\frac{1}{f} - 1)} \quad (23)$$

with

$$m^2 = \frac{\alpha}{e k_+ k_-}$$

In these equations $k_+$ and $k_-$ are the mobilities of the positive and negative charge carriers and $\alpha$ is the volume recombination constant (we used $k_+ = k_- = 3.2 \cdot 10^{-8} \text{ m}^2\text{V}^{-1}\text{s}^{-1}$ and $\alpha = 5.9 \cdot 10^{-16} \text{ m}^3\text{s}^{-1}$)\(^5\). Introducing the numerical data of the experimental set-up in equation 23 we obtain $f \geq 0.99$ for $V \geq 625\text{V}$. Then we only apply equation (22) to data for $V \geq 600 \text{V}$.

For lower voltages the free ion yield has to be extrapolated because volume recombination and space charge effects provoke charge losses and field screening, and equations (21) and (22) no longer hold. We expect from section 2.1 a linear relationship between the free ion yield and the electric field at low electric field values. Figure 4 shows this linear relationship between 600 V and 1000 V. At higher voltages the relationship begins to deviate from linearity as shows the figure. Then, we use data in the range $600 \text{V} \leq V \leq 1000 \text{V}$ to extrapolate the free ion yield at low electric field region.

\(^5\) Values obtained under irradiation with X rays, in agreement with [12].
This experimental linear relationship between free ion yield and external electric field $E$, is

$$G_{fi}(E) = (0.32 \pm 0.02) + (1.73 \pm 0.09) \cdot 10^{-7} \cdot E$$  \hspace{1cm} (24)$$

where $E$ is given in V/m and the free ion yield in pairs/100 eV. Equation (24) is valid up to $E = 1.2$ MV/m with a confidence level of 96%. For higher values be have to take into account more terms in the equation (6).

For isooctane the total number of electron-ion pairs produced per 100 eV of absorbed energy is $N_{tot} = 1.83$ (also calculated with the EGSnrc code). Inserting this value and the obtained $G_{fi}^{0}$ in equation (18) we can obtain the parameter $\rho$, for the delta (11), Gaussian type (12) and exponential (13) distribution functions. The results are $\rho = 168 \pm 6$ Å for first and second distributions, and $\rho = 178 \pm 10$ Å for the exponential one.

Taking the numeric values for $N_{tot}$ and $\rho$ we can calculate the theoretical prediction for free ion yield within the Onsager theory using equations (17), (14) and (2), and compare these theoretical results with experimental data. Figure 5 shows results obtained using the three considered distribution functions. Delta and Gaussian type distributions agree with the experimental data, but not the exponential distribution.

The intercept-to-slope ratio from equation (24) is $E_c = (1.83 \pm 0.12)$ MV/m. Within the Onsager framework we obtain (see equation (16) and figure 2), $E_c = E_0 = (1.74 \pm 0.02)$ MV/m at the room temperature, $T = (294 \pm 2)$ K.
Fig. 5. Free ion yield against external electric field in the linear region. The diamond points show the experimental points for $E \geq 0.75$ MV/m (for lower fields we have to extrapolate using equation (24)). The dotted, dashed and continuous lines correspond to the theoretical prediction using exponential, delta and Gaussian type distribution functions respectively.

Experimental value and theoretical value for $E_c$ are clearly in agreement.

4 Conclusions

We have obtained and analyzed data on isooctane free ion yield irradiated by $\gamma$ photons, from a cobalt source, within the framework of the Onsager theory. Three distribution functions (describing separation distance between electron-ion pairs when thermalization is achieved) have been considered: a delta function, a Gaussian type function and an exponential function. The first and the second describe data correctly in the covered electric field range, but not the exponential function.

This fact means that free ion yield depends in a fundamental way on the choice of the distribution function, which is not predicted by the theory. The good agreement between the experimental data and the theoretical prediction using a delta or a Gaussian type distribution with a dispersion parameter $\sigma = 0.25 \cdot \rho$ seems to show that in this case $f(r)$ is a Gaussian type function with a small dispersion parameter.

If electron would suffer a large number of independent collisions before thermalization, a Gaussian distribution function for the thermalization distances will agree with the central limit theorem. The lack of data about the nonpolar
liquids cross sections makes difficult to obtain models describing the nature of $f(r)$. Some computer simulations have been made (see for example [13]) in this sense, however more theoretical and numerical work is needed in this area.

On the other hand, the theoretical prediction for the intercept-to-slope ratio $E_c$, is in agreement with the experimental value. This is an important result to check the Onsager theory because it does not depend on $f(r)$.

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