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Characterization of tetramethylsilane for liquid-filled ionization dosimeters: Ion mobilities, free-ion yield and general recombination

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ABSTRACT

Liquid-filled ionization chambers (LICs) are interesting detectors for the dosimetry of radiotherapy beams due to their water-equivalent response and high spatial resolution. Isooctane is the liquid most often used as an active medium, but other hydrocarbons, particularly tetramethylsilane (TMS), can be suitable for dosimetry.

In this work we present a characterization of TMS (Merck, NMR calibration grade, purity > 99.7%) for its use in LICs. The characterization consisted of measuring ion mobilities, using low dose 6 MV photon pulses from a medical linac, and free-ion yield, using a continuous cobalt-60 beam (the reference beam quality used in radiotherapy dosimetry). Those values were then used to model general recombination in a TMS-filled LIC.

Measured ion mobilities, $(2.6 \pm 0.3) \times 10^{-8}$ and $(3.6 \pm 0.4) \times 10^{-8}$ $m^2\,V^{-1}\,s^{-1}$, are similar to mobilities in isooctane, and two- to three-fold lower than some values reported for TMS. Such discrepancy can probably be attributed to the presence of different impurities. On the other hand, free-ion yield values obtained are approximately two-fold higher than for isooctane, in agreement with published data. Such high free-ion yield values result in a higher signal-to-noise ratio and may allow even better spatial resolution to be obtained with TMS-filled LICs. However, it comes at the cost of higher recombination effects that can compromise the operation of the chamber. Such high recombination and the low boiling point of TMS ($\simeq 28$ °C) make isooctane-filled LICs preferable to TMS-filled LICS for radiotherapy applications.

1. Introduction

Liquid-filled ionization chambers (LICs) were introduced by Wickman for dosimetry in radiotherapy [1]. Research in LICs for radiotherapy has grown in the last two decades due to the good characteristics that they present for dosimetry, namely a near water-equivalent response, and the attainable high spatial resolution due to the high density of the ionization medium (when compared to air), which is especially important in the verification of small and/or high gradient fields such as those present in many advanced radiotherapy techniques. Several prototypes and commercial devices, as well as dosimetry methods, have been developed and characterized [1–11]. Isooctane (2,2,4-trimethylpentane, C_8H_{18}) is the liquid most commonly used in LICs due to its good physico-chemical properties. However, other hydrocarbons, especially tetramethylsilane (TMS, SiC_4H_{12}), have been used and may be adequate for dosimetry.

The main limitation of LICs is the large recombination effects they present due to low charge mobilities and high charge densities at standard therapy dose rates, which can potentially limit their application if not properly corrected for. Liquids employed as active media in LICs are not ultrapurified, which causes a suppression of electronic mobilities as negative charge is carried by electronegative impurities present in the liquid. High mass density of the media causes the thermalization of ionized electrons nearby ionized molecules, and a very fast recombination of an important fraction of the generated ion pairs occurs, a mechanism known as initial recombination [12]. The number of ions pairs escaping from initial

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recombination per 100 eV of deposited energy is known as free-ion yield, G_{fi}.

In this work we present a characterization of TMS (Merck, NMR calibration grade, purity $>99.7\%$) for its use as an active medium in LICs. We have measured ion mobilities, by studying the shape of the readout signal after a pulse of radiation, and free-ion yield, by analyzing the readout signal under continuous cobalt-60 irradiation. We have used those values to study general recombination in TMS-filled LICs at radiotherapy dose rates.

2. Materials and methods

2.1. Liquid-filled ionization chamber

The LIC used in this work is a detector array of 128 pixels arranged on a 16×8 grid. Each pixel has an effective area of $2\,mm\times2\,mm$ and a nominal gap of 0.5 mm. The readout is performed with the X-ray Data Acquisition System (Sens-Tech Ltd, UK), which has a sensitivity around 4200 pC/ADC and tunable integration time ranging from 0.01 ms to 0.5 s. More information on the design and characterization of this device (with isooctane as sensitive medium) can be found elsewhere [10]. The liquid used in this study is tetramethylsilane (Merck, NMR calibration grade) with a nominal purity $>99.7\%$.

2.2. Mobilities: experimental setup, data acquisition and analysis

The mobilities of the charge carriers were obtained by analyzing the shape of the readout signal after an ionization pulse. We used a 6 MV pulsed photon beam generated by a Siemens Mevatron LINAC. The pulse duration, a few μs, is much shorter than the typical charge collection time (a few ms), and thus does not perturb the shape of the readout signal. The LIC was placed at a 3 cm depth in a solid water phantom. We set the pulse repetition frequency to the minimum value ($\simeq 64$ Hz) in order to avoid overlapping between charge carriers ionized by consecutive pulses. The dose-per-pulse in the detector was around 0.06 mGy pulse^{-1}, which is low enough to guarantee a very high charge collection efficiency, thus avoiding the perturbation that general recombination could introduce in the pulse shape.

Two operation voltages were used, namely 1000 V and 1250 V. The integration time of the XDAS was set to 0.8 ms, providing an adequate balance between discretization of the readout signal and signal-to-noise ratio. The start of the acquisition was synchronized with the pulse via a trigger signal provided to the electronics after discrimination of console linac beam current signal. The shape of the readout signal was acquired for several thousand pulses per voltage value. The mobilities were then obtained by studying the temporal profile of the collection signal, including the effect of integration, following the method presented in [13].

The associated uncertainties, given with coverage factor $k=1$, were obtained as the combination of type A (standard deviation of the results for the number of pulses analyzed) and the type B uncertainty associated with the gap of the detector. The gap was determined by capacitance measurements to be 0.508 ± 0.023 mm.

2.3. Free ion yield: experimental setup, data acquisition and analysis

Free-ion yield values, G_{fi}, were obtained from measurements performed in a continuous cobalt-60 beam, the reference beam quality used in radiotherapy dosimetry. The dose rate at the LIC position was measured with a reference chamber to be $\dot{D}=0.339$ Gy min^{-1}, with a 0.5% relative uncertainty, but due to the perturbation effect caused by the non-water equivalence of the LIC (calculated with Monte Carlo) the dose rate on the LIC becomes 0.361 ± 0.004 Gy min^{-1}. The detector was polarized at 750 V, 1000 V, 1250 V and 1500 V, and the integration time of the readout electronics was 10 ms. At this dose rate and polarization voltages, general recombination is negligible and does not affect the determination of the free-ion yield. The temperature during the irradiations was 20 °C.

The G_{fi} is obtained from the readout signal S is

$$G_{fi}=S\frac{6000}{\dot{D}\rho t_{int}AhC} \tag{1}$$

where S is measured in ADC counts (and averaged over a few hundred acquisitions in order to obtain a reliable mean value and associated uncertainty), C is the sensitivity of the readout electronics in ADC/C units, t_{int} is the integration time of the readout electronics in seconds, ρ is the mass density of the liquid (648 kg m^{-3}), and $A\times h$ is the volume of the pixel in m^3. The factor 6000 originates from the conversion of minutes (dose rate) to seconds (integration time), and eV to 100 eV.

The uncertainties of the G_{fi} values can be obtained from the above expression by using standard uncertainty propagation laws, and are dominated by the uncertainty of the gap: $u(S)/S\sim0.4\%$, $u(C)/C\sim2\%$, $u(h)/h\sim4.5\%$, $u(\dot{D})/\dot{D}\sim1\%$.

2.4. General recombination

Radiotherapy is mostly performed with pulsed radiation beams. General collection efficiencies, f, in LICs irradiated with pulsed radiation beams are well described in the near saturation range $f>0.9$ by the Boag equation [14–16,8]:

$$f=\frac{1}{u}\log(1+u) \tag{2}$$

$$u=\frac{\alpha r h^2}{eV(k_1+k_2)} \tag{3}$$

where h is the detector gap, V the operation voltage, k_1 and k_2 the mobilities of positive and negative charge carriers, r the ionized charge per unit volume and pulse (after initial recombination), α the general recombination constant and e the electron charge. The recombination constant is given by the Debye equation [15], which cancels out the dependency of recombination on the mobilities:

$$\alpha=\frac{e}{\epsilon}(k_1+k_2) \tag{4}$$

where ϵ is the dielectric constant of the liquid ($1.92\epsilon_0$ for TMS).

The Boag equation assumes no overlapping of charge ionized by consecutive pulses, which would increase general recombination. Thus, the radiation pulse period p must be larger than the charge collection time:

$$t_{col}=\frac{h^2}{V\min(k_1,k_2)}<p. \tag{5}$$

We have computed collection efficiencies with these models, using the parameters obtained in this work, in order to investigate general recombination in TMS-filled LICs at therapeutic dose rates. We have also compared the reported experimental collection efficiencies with the values obtained here.

3. Results and discussion

3.1. Ion mobilities

Fig. 1 shows the time shape of two readout signals for 1000 and 1250 V, and the fit of the experimental data to the collection model. Table 1 shows the mobilities obtained for both operation voltages. The associated uncertainties are roughly equi-split between Type A (standard deviation of fits to many pulses) and Type B uncertainties