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2 **On the property of measurements with the PTW microLion** 3 **chamber in continuous beams**

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16 **Purpose:** The performance of liquid ionization chambers, which may prove to be useful
17 tools in the field of radiation dosimetry, is based on several chamber and liquid specific
18 characteristics. The present work investigates the performance of the PTW microLion
19 liquid ionization chamber with respect to recombination losses and perturbations from
20 ambient electric fields at various dose rates in continuous beams.

21 **Methods:** In the investigation, experiments were performed using two microLion
22 chambers, containing isooctane (C_8H_{18}) and tetramethylsilane ($Si(CH_3)_4$) as the
23 sensitive media, and a NACP-02 monitor chamber. An initial activity of approximately
24 250 GBq ^{18}F was employed as the radiation source in the experiments. The initial dose
25 rate in each measurement series was estimated to 1.0 Gy min^{-1} by Monte Carlo
26 simulations and the measurements were carried out during the decay of the radioactive
27 source. In the investigation of general recombination losses, employing the two-dose-
28 rate method for continuous beams, the liquid ionization chambers were operated at
29 polarizing voltages 25, 50, 100, 150, 200 and 300 V. Furthermore, measurements were
30 also performed at 500 V polarizing voltage in the investigation of the sensitivity of the
31 microLion chamber to ambient electric fields.

Results: The measurement results from the liquid ionization chambers, corrected for general recombination losses according to the two-dose-rate method for continuous beams, had a good agreement with the signal to dose linearity from the NACP-02 monitor chamber for general collection efficiencies above 70%. The results also displayed an agreement with the theoretical collection efficiencies according to the Greening theory, except for the liquid ionization chamber containing isoctane operated at 25 V. At lower dose rates, perturbations from ambient electric fields were found in the microLion chamber measurement results. Due to the perturbations, measurement results below an estimated dose rate of 0.2 Gy min^{-1} were excluded from the present investigation of the general collection efficiency. The perturbations were found to be more pronounced when the chamber polarizing voltage was increased.

Conclusions: By using the two-dose-rate method for continuous beams, comparable corrected ionization currents from experiments in low- and medium energy photon beams can be achieved. However, the valid range of general collection efficiencies has been found to vary in a comparison between experiments performed in continuous beams of 120 kVp x-ray, and the present investigation of 511 keV annihilation photons. At very high dose rates in continuous beams, there are presently no methods that can be used to correct for general recombination losses and at low dose rates the microLion chamber may be perturbed by ambient electric fields. Increasing the chamber polarizing voltage, which diminishes the general recombination effect, was found to increase the microLion chamber sensitivity to ambient electric fields. Prudence is thus advised when employing the microLion chamber in radiation dosimetry, as ambient electric fields of the strength observed in the present work may be found in many common situations. Due to uncertainties in the theoretical basis for recombination losses in liquids, further studies on the underlying theories for the initial and general recombination effect are needed if liquid ionization chambers are to become a viable option in high precision radiation dosimetry.

Keywords: General recombination, continuous beam, two-dose-rate method, liquid ionization chamber, radiation dosimetry, isooctane, tetramethylsilane, perturbation, ambient electric field

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I. INTRODUCTION

During recent years there has been an increased interest in the use of liquid ionization chambers (LICs) in radiation dosimetry applications.¹⁻⁴ The density of the sensitive media employed in LICs gives a much higher sensitivity than gas-filled ionization chambers. This property is desirable in certain dosimetric applications since LICs can be manufactured with small dimensions and thus making them suitable for applications requiring high spatial resolution. Such applications are found both in radiation therapy and diagnostics.⁵⁻⁹ However, due to the relatively high density of a liquid and low ion drift velocity the recombination losses in LICs are substantial, especially at high dose rates. Since recombination losses yield non-linear effects, the interpretation of the measured current or charge with a LIC is not straightforward. Much effort has therefore been given in finding experimental methods that accurately correct for general recombination losses.¹⁰⁻¹⁴

The recent work by Andersson and Tölli¹⁴ involves an application of the two-dose-rate method for LICs in continuous beams. The theory by Greening¹⁵ was used as a model for general recombination losses in the liquids. The authors showed that the method works well in achieving a similar signal to dose relationship in a LIC as compared to an air-filled reference ionization chamber in continuous beams for general collection efficiencies above 90%. The results were also in good agreement with the theoretical collection efficiency according to the Greening equation over the same interval of general collection efficiencies. The validity of the two-dose-rate method was investigated by experiments in a continuous x-ray beam with a tube potential of 120 kV.

Ionic recombination effects, and methods that accurately correct for the associated losses of measurement signal, are not the only factors that determines the performance of LICs in radiation dosimetry. There are several chamber and liquid specific characteristics that can have an impact on the usability of LICs, these include particle type and energy dependence, temperature dependence and

leakage currents. An investigation of these effects in LICs can be found in Wickman *et al.*¹⁶ Leakage currents set a lower limit for permissible dose rates that may be accurately measured by LICs, similarly to how recombination effects set an upper limit of dose rates that can be determined by measurements with LICs. In between these limits, the valid operational range of dose rates for LICs is found.

In the present work, an experimental investigation is carried out on the performance of the PTW microLion LIC at various dose rates in continuous beams of annihilation photons from the decay of the radioactive isotope ¹⁸F. In order to achieve accurate results from measurements performed at higher dose rates, the two-dose-rate method is employed to correct for general recombination losses. By using a decaying source in the experiments, it is furthermore possible to investigate the performance of the microLion chamber at low dose rates, down to the region where leakage currents should be the limiting factor. In the present work we have thus chosen to study how the performance of the microLion chamber is affected when used in environments with ambient electric fields. The two-dose-rate method was experimentally shown to be independent of the initial recombination effect in the work by Andersson and Tölli,¹⁴ in which measurements were performed at several different LIC polarizing voltages. However, the initial recombination effect may not depend only on the polarizing voltage but also on radiation beam type and energy, as the radiation quality will influence the density of ions created along an incident ionizing particle track. Wickman *et al.*¹⁶ have reported evidence of energy dependence for LICs, which may influence the initial recombination effect in liquids, in the low- to medium photon energy regime. Since the two-dose-rate method does not explicitly model initial recombination, the present work thus includes a further analysis of the properties of two-dose-rate method, based on measurements of 511 keV annihilation photons and the results from measurements of 120 kVp x-ray photons by Andersson and Tölli.¹⁴

II. MATERIALS

II.A. Ionization chambers

Two plane parallel and sealed LICs (microLion, PTW, Germany), containing isooctane (C_8H_{18}) and tetramethylsilane ($Si(CH_3)_4$), were used in the experiments. The LICs are manufactured to be geometrically identical with radius 1.25 mm and an electrode separation of 0.35 mm. Additionally, an air-filled NACP-02 plane parallel ionization chamber (s/n 104), built and designed at the Laboratory of Radiation Physics, Umeå University, has been used as a monitor chamber. The NACP-02 chamber has a measurement volume diameter of 10 mm and an electrode separation of 2 mm.

II.B. Electrometers and high voltage supplies

The charge from the ionization chambers was collected and read out by electrometers of the types UNIDOS and UNIDOS Atto (PTW, Germany) for the NACP-02 chamber and the LICs, respectively. The NACP-02 chamber polarizing voltage was supplied by the UNIDOS electrometer while a Keithley Model 248 High Voltage Unit supplied the polarizing voltage for the LICs. The experiments were performed with the LICs operated at polarizing voltages 25, 50, 100, 150, 200, 300 and 500 V, which gives electric field strengths between 0.07 and 1.4 MV m⁻¹, over the chamber effective measurement volume. The measurements at 500 V polarizing voltage were only employed in the investigation of perturbations from ambient electric fields. The NACP-02 chamber was operated at a polarizing voltage of 400 V.

II.C. Irradiation source and geometry

In the present experiments, 511 keV annihilation photons following the decay of the radioactive isotope ^{18}F (half-life 109.77 minutes¹⁷) were employed. A cyclotron (GE Medical, PETtrace 6) at the University hospital of Norrland (Umeå, Sweden) was employed to produce the isotopes used in the experiments. Each measurement series started with an activity of approximately 250 GBq of ^{18}F , which is the result of a 120 minutes production time at a cyclotron current of 60 μA , as specified by the manufacturer. The activity, which is contained in liquid form (2 ml), was transported by high pressure helium gas to a hot cell in the PET radiation chemistry laboratory at the hospital. The hot cell

(BBS2-V, Comecer S.p.A), which has the inner dimensions 1031 x 1024 x 894 mm³ (height, width and depth), is shielded by 75 mm lead clad with stainless steel. The measurement geometry for the ionization chambers in relation to the radiation source within the hot cell is shown in Fig. 1.

FIG. 1. Left: The measurement geometry and experimental setup used for the experiments with ¹⁸F. Middle, Right: Schematic and exploded view drawing of the experimental setup, showing the LIC and NACP-02 chamber configuration in relation to the glass vial container for the activity of ¹⁸F.

In the measurement geometry, the glass vial container (5 ml) for the activity of ¹⁸F was fixed in a perspex holder. The perspex holder, which was fitted in contact with the NACP-02 chamber front wall, had a thickness of 6 mm to achieve a suitable build-up for the 511 keV photons resulting from the positron-electron annihilation. The LICs were placed in such a manner that the front wall was in contact with the back wall of the NACP-02 chamber inside a perspex holder, as shown in Fig. 1. The ionization chambers and the glass vial containing the activity were held in place on a perspex stand, which placed the radiation source in the geometrical center of the hot cell. The temperature in the hot cell was monitored and regulated to 20 °C (temperature sensor accuracy better than ±0.5 °C, as specified by the manufacturer), and the air pressure was monitored and regulated relative to the environment outside the laboratory to ensure that radioactive isotopes or other harmful substances cannot migrate to surrounding areas.

The production of ¹⁸F is achieved by irradiating 16.5 MeV protons, produced in the cyclotron, on a target volume containing enriched ¹⁸O water. Due to impurities in the target, most commonly from the cyclotron rinsing water, a certain amount of other isotopes is also produced. The most frequently

occurring isotope in this regard is ^{13}N (half-life 9.97 minutes¹⁷), which is a positron emitter. Approximately 30 minutes after delivery to the hot cell (three half-lives of ^{13}N), the amount of ^{13}N or other contaminant isotopes remaining in the batch of ^{18}F may thus be considered negligible because of short half-lives and a relatively low yield. A study of the present cyclotron technology and radionuclide impurities in water samples irradiated in a niobium target has been given by Avila-Rodriguez *et al.*¹⁸ The total amount of gamma-emitting radioactive impurities has been found to be in the order of 15 kBq when the target was irradiated with 16.5 MeV protons during 90 minutes at a cyclotron current of 50 μA .¹⁹ These cyclotron settings results in a batch of approximately 150 GBq ^{18}F , as compared to the present work where batches of 250 GBq ^{18}F were used.

III. METHODS

III.A. Theory and experiments

The two-dose-rate method for continuous beams by Andersson and Tölli¹⁴ is based on the Greening equation¹⁵, which is given by

$$f = \frac{1}{1 + \frac{1}{6}\xi^2}, \text{ where } \xi^2 = m^2 \frac{h^4 q_0}{U^2}, \text{ and } m = \left(\frac{\alpha}{ek_1 k_2} \right)^{1/2}. \quad (1)$$

The parameters above have the following meanings, h is the separation between the collecting electrodes, U is the chamber polarizing voltage, k_1 and k_2 are the mobilities of the positive and negative ions, α is the general recombination rate constant, q_0 is the charge per unit volume and unit time escaping initial recombination and e is the elementary charge.

The two-dose-rate method for continuous beams uses measured values at a set of two dose rates, d_1 and d_2 , to determine $\xi^2(d_1)$ from

$$\xi^2(d_1) = \frac{\left(\frac{Q_{NACP}(d_1)}{Q_{NACP}(d_2)} - \frac{Q_{LIC}(d_1)}{Q_{LIC}(d_2)} \right)}{\frac{1}{6} \left(\frac{Q_{LIC}(d_1)}{Q_{LIC}(d_2)} - 1 \right)}. \quad (2)$$

Here, $Q_{NACP}(d_i)$ and $Q_{LIC}(d_i)$ are the charges measured with a NACP-02 monitor chamber and a LIC at different dose rates d_i ($i=1,2$). Thus, in the two-dose-rate method, $\xi^2(d_1)$ is determined entirely experimentally and the collection efficiency with respect to general recombination at dose rate d_1 in a LIC can then be calculated using $\xi^2(d_1)$ in Eq. 1. For details of the derivation of Eq. 2, see Andersson and Tölli.¹⁴ The air-filled NACP-02 chamber has a well-known pressure and temperature dependence and can readily be corrected for its own general recombination losses by the two-voltage method.

Due to the radioactive source decay in the present experiments, we may also express $\xi^2(d_1)$ as

$$\xi^2(d_1) = \frac{\left(e^{\lambda(t_2-t_1)} - \frac{Q_{LIC}(d_1)}{Q_{LIC}(d_2)} \right)}{\frac{1}{6} \left(\frac{Q_{LIC}(d_1)}{Q_{LIC}(d_2)} - 1 \right)}, \quad (3)$$

where λ is the decay constant and t_i corresponds to the time at which the dose rate is equal to d_i , thus eliminating the need of a reference ionization chamber. To investigate the two-dose-rate method for LICs in continuous beams from a decaying radioactive source several experiments were carried out, where the LIC voltage was kept constant for each batch of ^{18}F . The collection and recording of measurement results from the electrometers during the decay of the radioactive source was controlled by a computer program developed in LabView (Full Development System version 8.6, National Instruments). Each measurement series, for a given batch of ^{18}F , was divided into measurement cycles of 60 s, and the charge was read out and recorded after each cycle for the LICs and the NACP-02 chamber, respectively. The general collection efficiencies for the LICs were determined by using different combinations of the results from each measurement cycle representing various dose rates (d_i), recorded during the decay of the radioactive source, in Eq. 2 and 3.

III.B. Monte Carlo simulation

The dose rates, in which the present experiments were carried out, were determined by means of Monte Carlo simulation using Visual Editor v. X_24b that runs MCNPX v. 2.7c. The geometry used in

the simulations was designed in accordance with Fig. 1, and the geometrical data used for the LICs was given by PTW (personal communication). The positron spectra representing the decay of ^{18}F were taken from Chu *et al.*¹⁷

The results from the Monte Carlo simulations were given as absorbed dose in the LICs per emitted positron. From these results, the dose rates in the LICs were calculated using the production estimate of 250 GBq ^{18}F , as specified by the manufacturer. This gave an estimated maximum dose rate of 1.0 Gy min⁻¹ to the LIC effective measurement volume for the present experimental setup and method.

IV. RESULTS

The recording of measurement results from each batch of ^{18}F was started 30 minutes after the activity had been delivered to the hot cell to avoid measurement signals from isotopes other than ^{18}F , as described in Sec. II and III. Variations in the cyclotron yield were found for the different experiments by studying the NACP-02 chamber measurement results, the initial activity delivered to the hot cell had a relative standard deviation of 3.4%. The NACP-02 chamber measurement results were therefore used to correlate the different measurement series to that with the lowest cyclotron yield. By this handling of the measurement results, and without interpolating in each measurement series, the initial activity used in the calculations of the general collection efficiencies had a relative standard deviation of 0.2% for the different experiments.

The leakage currents in the LICs were recorded before each experiment and the measurement results were corrected by the mean value of the respective leakage currents. For the different experiments, the leakage currents were found to be of both positive and negative sign, for both liquids, and the maximum value was 27 fA for isooctane and 101 fA for tetramethylsilane. The maximum leakage currents amounts to 0.4 and 1.0% of the ionization currents at the lowest dose rates and polarizing voltages employed in this work for both liquids, respectively.

At low dose rates, as well as in measurements of leakage currents, periodic disturbances in the measurement signal from the LICs were observed. As these disturbances will perturb the results from

the two-dose-rate method, measurement results below an estimated dose rate of 0.2 Gy min^{-1} have not been used for the calculations of recombination losses in this work. The limit was conservatively chosen as it corresponds to ionization currents that are at least a factor two higher than those at which the perturbations were evident in the measurement results. In a comparison between the temperature variations recorded by the clean room monitoring system, which manages the laboratory environment, and the perturbations in the measurement results a similar periodicity was observed. An additional experiment was performed with the LIC containing isooctane operated at 500 V, since the chamber polarizing voltages used in this work for the determination of general collection efficiencies are below those recommended by the manufacturer. At this higher polarizing voltage, the perturbations were even more pronounced, becoming evident at higher dose rates. An example of the measurement results from the LIC containing isooctane and the NACP-02 chamber, the perturbations and the temperature variations are given in Fig. 2 and 3 (Top row). The LIC and NACP-02 measurement results are well correlated in time since a LabView program handled the data recording, as described in Sec. III.A. Similarly, the recording of temperature variations by the clean room monitoring system was also computerized. The comparisons in Fig. 3 (Top row) were thus made by correlating the time stamps from the different computer systems.

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258 FIG. 2. Measurement results for the LIC containing isooctane operated at 300 and 500 V chamber
259 polarizing voltage, and the NACP-02 chamber in the hot cell. The results for the LICs and the NACP-
260 02 chamber are indicated by solid and dashed lines, respectively. Figures on the right hand side are
261 showing the LIC perturbations on a more detailed scale.

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FIG. 3. Top row: Measurement results from the LIC containing isooctane operated at 300 and 500 V chamber polarizing voltage, and the recorded temperature variations in the hot cell. The measurement results from the LIC and the temperature variations are indicated by thick and thin lines, respectively. Bottom row: Variations of the electric field in the hot cell, as measured by the C.A 42 instrument, and manually correlated examples of the LIC measurement results for 300 and 500 V chamber polarizing voltage. Measurements of the ambient electric fields were performed separately from the ion chamber experiments. The measurement results from the LIC and the ambient electric field variations are indicated by solid thick lines and dotted lines, respectively.

To investigate the LIC perturbations in regard to possible influences from ambient electromagnetic fields, measurements of the extremely low frequency (ELF) field strength variations in the hot cell were performed with an electric field probe (C.A 42 EF400, Chauvin Arnoux, France) and a magnetic field recorder (EMDEX II, Enertech, USA). The measurements of the magnetic flux density did not yield any results with a variation in time corresponding to the perturbations. The measurement results from the electric field probe, in the frequency range 5 Hz – 3.2 kHz, displayed a variation in time that corresponded to the perturbations. The electric field probe was battery-powered, and measurements of electric ELF fields were thus limited in time to approximately 80 minutes. Since the perturbations investigated had a period of approximately 35 minutes, several measurements with the electric field

probe were performed to quantify the ambient electric field variations. An example of the measured ambient electric fields and the LIC perturbations for the LIC containing isooctane operated at 300 and 500 V are given in Fig. 3 (Bottom row). The measurements with the electric field probe were performed separately from the experiments with ^{18}F , and the perturbations in the LIC measurement results and the measured ambient electric fields were manually correlated peak to peak to show the agreement in periodicity in Fig. 3 (Bottom row). The variations of the ambient electric field strength that correlated to the perturbations were approximately in the range $100 - 250 \text{ V m}^{-1}$. Since the LIC perturbations were found to correlate to both the variations in temperature and the ambient electric field, it is likely that the regulatory system for the temperature in the PET radiation chemistry laboratory was the source of these ambient electric fields. The measurement results of the ambient electric field strength variations also included lower readings (much smaller than 100 V m^{-1}), which did not have a correlation to any perturbations found in the LIC measurement results and the source of these lesser ambient field variations is not known. The measurement setup was also moved to a different hot cell, in another part of the PET radiation chemistry laboratory, where the ambient electric field levels were below the measurement range of the electric field probe (1 V m^{-1}). This gave measurement results from the LICs without perturbations, even at low dose rates. No perturbations in the measurement results from the NACP-02 chamber were found in either of the hot cells.

By sampling the signal from the ionization chambers during the decay of the radiation source, $Q_{\text{NACP}}(d_i)$ and $Q_{\text{LIC}}(d_i)$ were recorded by the LabView program, and several hundred combinations of measurement values at various dose rates between 0.2 and 1.0 Gy min^{-1} were employed to determine each $\xi^2(d_i)$ in steps of 0.1 Gy min^{-1} . General collection efficiencies resulting from calculations performed according to the two-dose-rate method, utilizing the NACP-02 chamber according to Eq. 2 are presented in Table I and II for isooctane and tetramethylsilane, respectively. General collection efficiencies, for which the ratios of the NACP-02 readings were replaced by the expression based on exponential decay (Eq. 3), displayed a maximum deviation of 0.8% from the results calculated according to Eq. 2.

TABLE I. General collection efficiencies, f , determined according to the two-dose-rate method for the LIC containing isooctane at different polarizing voltages and dose rates. Values in the parenthesis represent one relative standard deviation expressed as a percentage.

Dose rate (Gy min ⁻¹)	f (25 V)	f (50 V)	f (100 V)	f (150 V)	f (200 V)	f (300 V)
0.2	0.942 (0.1)	0.981 (0.1)	0.994 (0.1)	0.996 (0.1)	0.998 (0.1)	1.000 (0.1)
0.4	0.875 (0.2)	0.961 (0.2)	0.985 (0.3)	0.995 (0.2)	0.993 (0.1)	1.000 (0.2)
0.6	0.815 (0.1)	0.937 (0.1)	0.980 (0.2)	0.991 (0.1)	0.995 (0.1)	0.999 (0.1)
0.8	0.780 (0.4)	0.921 (0.1)	0.976 (0.1)	0.988 (0.1)	0.994 (0.1)	1.000 (0.1)
1.0	0.737 (0.5)	0.900 (0.1)	0.970 (0.1)	0.986 (0.1)	0.992 (0.1)	1.000 (0.1)

TABLE II. General collection efficiencies, f , determined according to the two-dose-rate method for the LIC containing tetramethylsilane at different polarizing voltages and dose rates. Values in the parenthesis represent one relative standard deviation expressed as a percentage.

Dose rate (Gy min ⁻¹)	f (25 V)	f (50 V)	f (100 V)	f (150 V)	f (200 V)	f (300 V)
0.2	0.950 (0.1)	0.983 (0.1)	0.994 (0.1)	1.000 (0.04)	0.996 (0.2)	1.000 (0.1)
0.4	0.893 (0.2)	0.962 (0.1)	0.983 (0.5)	0.999 (0.1)	0.992 (0.2)	0.999 (0.3)
0.6	0.839 (0.2)	0.945 (0.1)	0.979 (0.3)	0.996 (0.1)	0.992 (0.2)	0.995 (0.1)
0.8	0.808 (0.3)	0.929 (0.1)	0.975 (0.3)	0.989 (0.2)	0.992 (0.1)	0.994 (0.1)
1.0	0.767 (0.3)	0.905 (0.1)	0.970 (0.2)	0.979 (0.3)	0.990 (0.1)	0.991 (0.1)

In order to investigate the accuracy of the corrections made by the two-dose-rate method on the LIC measurement values, the corrected LIC to NACP-02 chamber ratios were calculated for the different dose rates and chamber polarizing voltages used in this work. These ratios are shown in Fig. 4 for both isooctane and tetramethylsilane.

FIG. 4. Corrected LIC to NACP-02 ratios for isooctane and tetramethylsilane for the LIC polarizing voltages and dose rates used in the present work. The dashed line indicates the mean LIC to NACP-02 chamber ratio. The corrected LIC to NACP-02 ratios are given with one standard deviation.

The corrected LIC to NACP-02 chamber ratios for the present dose rates, per LIC polarizing voltage, had relative standard deviations below 0.4% for both liquids. The relative standard deviation was largest for the lowest chamber polarizing voltage for both isooctane and tetramethylsilane. Furthermore, the LIC containing isooctane displayed a greater stability in measurement results than the LIC containing tetramethylsilane, as seen from the standard deviations in Fig. 4. For isooctane, with the exception of 25 V, the relative standard deviations per chamber polarizing voltage were below 0.1%.

The theoretical collection efficiency, as proposed by Greening,¹⁵ can be calculated directly from the LIC measurement results, as shown by Andersson and Tölli.¹⁴ A comparison between experimental and theoretical general collection efficiencies is shown in Fig. 5 for both isooctane and tetramethylsilane. The parameters employed in the calculations of the theoretical general collection efficiencies were taken from Johansson and Wickman¹⁰ for the liquids (ion mobilities and recombination coefficients), and the LIC manufacturer chamber dimension specifications.

FIG. 5. A comparison of the theoretical collection efficiency according to Greening (▽) and the experimental collection efficiency according to the two-dose-rate method (○) for isooctane and tetramethylsilane at the different LIC polarizing voltages and dose rates in the present work.

The experimental and theoretical general collection efficiencies are in good agreement for isooctane for all polarizing voltages, except 25 V. Here, the deviation between experiment and theory is up to 8% for the dose rate 1.0 Gy min^{-1} , where the theoretical collection efficiency is calculated to 0.685. In general, the experimental and theoretical collection efficiencies are in better agreement for tetramethylsilane than for isooctane except for a few cases, which have no systematic relation to dose rate or LIC polarizing voltage. This may be attributed to the somewhat lesser stability of the LIC containing tetramethylsilane, as previously noted. For example, there are deviations up to 2% for 50 V and 0.9% for 150 V for the LIC containing tetramethylsilane.

V. DISCUSSION

Two phenomena with a temporal variation similar to the LIC perturbations were observed in the PET radiation chemistry laboratory. These were the periodic variations in temperature and ambient electric field strength. The maximum temperature variations in the measurement series shown in Fig. 3 (Top row) are approximately 0.1 and 0.4 °C for 300 and 500 V chamber polarizing voltage, respectively as measured by the clean room monitoring system. Wickman *et al.*¹⁶ have reported a temperature dependence of 0.4 and 0.3% °C⁻¹ for LICs filled with isooctane and tetramethylsilane, respectively. Furthermore, Franco *et al.*²⁰ have investigated the temperature dependence of a LIC containing isooctane in relation to the chamber collection electric field, by employing the Onsager theory.²¹ By calculations according to the work by Franco *et al.*²⁰ the temperature dependence of isooctane for the chamber polarizing voltages used in the present work is between 0.3 and 0.6% °C⁻¹. From the findings of Wickman *et al.*¹⁶ and Franco *et al.*²⁰ the LIC measurement signal is expected to increase with increasing temperature while the opposite relation between temperature and signal due to the perturbation was found in the present work, as seen in Fig. 3 (Top row). The investigation by Franco *et al.*²⁰ also show that the temperature dependence of isooctane decreases with increasing chamber polarizing voltage, and the opposite relation was observed in the perturbations found in the present work, as noted in Sec. IV. The magnitudes of the temperature variations observed here are thus too small to have caused the perturbations, and the observed temperature variations and perturbations display a relationship that is the exact inverse to what should be expected according to what has been

reported.^{16,20} From these arguments it can be concluded that variation in temperature was not the cause of the perturbations in the present experiments.

The collection electric field strengths used for the LICs in the present work were between 0.07 and 2.4 MV m⁻¹, and the ambient electric field variations that were observed to correspond to the perturbations were approximately in the range 100 – 250 V m⁻¹. The possible perturbation effect, by direct superposition, on the collection electric field over the liquid layer from the ambient electric field strengths observed is thus at most 0.4% for the present LIC polarizing voltages. Perturbations of the magnitude observed here can thus not have originated in the effective measurement volume. As noted in Sec. IV, the experimental setup was also moved to a different hot cell, in another part of the PET radiation chemistry laboratory, where the electric field probe recorded no ambient electric fields. In this hot cell no perturbations were found in the LIC measurement signal, even at low dose rates. No perturbations were found in the NACP-02 chamber measurement results, in either of the hot cells. From these findings it can be concluded that the microLion chamber is sensitive to ambient electric fields, but the sensitivity cannot be traced to the liquid in the effective measurement volume nor the collection electric field applied over the liquid layer. The sensitivity must thus originate in another part or aspect of the microLion chamber. There are two probable sources for the sensitivity to ambient electric fields, which may be co-contributors to the perturbations observed in the present work. These are unshielded sections of the conduction strands inside the microLion chamber body, and that the chamber by design does not have guard electrodes for the effective measurement volume.

By fundamental electromagnetic theory it is known that for a dielectric material, such as the microLion chamber body, the presence of an ambient electric field ($\vec{E}$) will result in an electric displacement field ($\vec{D}$). The electric displacement field will in turn give a displacement current density ($\vec{J}_D$) in the material, which is the time-derivative of the displacement field $\left(\vec{J}_D = \frac{\partial \vec{D}}{\partial t}\right)$. A current in an unshielded conduction strand inside the chamber body will be susceptible to perturbations from displacement currents. To quantify the possible displacement currents in the

present experimental setup, detailed knowledge regarding the frequencies of the ambient electric fields would be required, as well as the exact construction of the microLion chamber and the orientation of the chamber in regard to the ambient electric field lines. None of these parameters are well known in the present work, and it is thus not possible to quantify the influence of displacement currents on conduction strands inside the microLion chamber body. As previously discussed, the microLion chamber effective measurement volume does not have guard electrodes. This means that the collection electric field will extend outside the effective measurement volume, to an extent depending on the LIC polarizing voltage. An ambient electric field will thus, by the resulting electric displacement field, interfere directly by superposition with extensions of the collection electric field present outside the effective measurement volume. Such interferences may in turn perturb the current in unshielded conduction strands and this is thus a plausible explanation of why the perturbations are observed to not only depend on ionization current but also on the LIC polarizing voltage.

The PET radiation chemistry laboratory is not well suited for further and more detailed experimental studies of the relationship between ambient electric fields, ionization currents and polarizing voltages, as the environmental variables are automatically controlled for optimized clinical operation. Further studies by measurements at a certified EMC laboratory and electrodynamics simulations are needed to investigate the microLion chamber susceptibility to ambient electric fields, and is thus beyond the scope of this investigation. In summary, from the experimental findings in the present work, the microLion chamber is sensitive to ambient electric fields and the sensitivity is likely related to an insufficient shielding of the chamber in conjunction with the absence of guard electrodes for the effective measurement volume. The LIC perturbations have been found to depend on the ambient electric field strength, ionization current and also chamber polarizing voltage. Furthermore, the perturbations have been observed to increase in strength and appear at higher dose rates with increased chamber polarizing voltage. Prudence is advised when employing the microLion chamber for purposes of radiation dosimetry as ambient electric fields, which have been observed to cause perturbations in measurement results, can be found in many common situations.

The ionization currents at which the perturbations from ambient electric fields were evident in the present experiments are very low compared to those found in the investigations by Tölle *et al.*¹³ and Andersson and Tölle.¹⁴ Furthermore, the investigation by Tölle *et al.*¹³ involved measurements in a water phantom, where no stability issues for the microLion chamber involving ambient electric fields have been reported. The problems encountered in the present measurements are thus unlikely to have caused any errors of significance in previously mentioned works.

The two-dose-rate method for general recombination correction for LICs in continuous beams that has been used in this work shares conceptual and practical traits with the widely used two-voltage method for air-filled ionization chambers. The experimental nature of the method allows for a determination of the collection efficiency with respect to general recombination without any presumed knowledge about parameters such as chamber dimensions, general recombination rate constants or ion mobilities. These parameters, which are required in theoretical calculations of the collection efficiency with respect to general recombination, can vary depending on the purity of the liquid and the accuracy of the LIC manufacturing process.

The NACP-02 chamber measurement results were not corrected for general recombination losses, as the dose rates in the present work are lower than those used by Andersson and Tölle¹⁴ where the general recombination losses were negligible ($< 0.1\%$). Since the temperature and pressure in the hot cell was monitored and regulated, as noted in Sec. II.C, no corrections were made to the NACP-02 chamber readings. To verify that the NACP-02 chamber was not perturbed by general recombination losses or variations in pressure and temperature, the measurement results were analyzed by exponential regression for each experiment to determine the half-life of ^{18}F . The result was a mean value of 109.73 minutes with a relative standard deviation of 0.1%, which is in close agreement with the value reported by Chu *et al.*¹⁷ (109.77 minutes). This shows that negative effects from general recombination, and variations in temperature and pressure on the NACP-02 chamber measurement results were negligible. The mean temperature in the hot cell, which was set to 20 °C in the clean room monitoring system, recorded during the experiments was in fact closer to 21 °C, as seen in Fig. 3 (Top

row). The maximum temperature variation recorded was 0.5 °C for all the different measurement series.

The initially delivered activity of ^{18}F had a relative standard deviation of 3.4% for the experiments in the present work, as determined by the NACP-02 chamber measurement results. Furthermore, due to possible trace levels of contaminants in the cyclotron target, a small amount of undesired isotopes may be produced in addition to the activity of ^{18}F . These isotopes and their respective half-lives are well known, and as previously discussed, waiting a period of 30 minutes after the activity has been delivered to the hot cell should result in negligible amounts of contaminant isotopes.^{18,19} The activity of ^{18}F produced by the cyclotron according to the settings in the present work was approximately 250 GBq, as specified by the manufacturer. Monte Carlo simulations based on the manufacturer specifications were employed to determine the dose rates in the present work. Due to this approach, an uncertainty in the dose rate estimates of the same magnitude as the variations observed for the cyclotron yield from the NACP-02 chamber measurement results is introduced. Additional uncertainty in the dose rate estimates may come from the positioning of the experimental setup in the hot cell, as well as from small variations in the relative placement of the ionization chambers and the vial containing the radiation source. Much effort was therefore given to reproduce the setup for each experiment, including placement in the hot cell and fixation of ionization chambers and the vial containing the activity in the arrangement of perspex holders shown in Fig. 1.

The experimentally determined collection efficiencies with respect to general recombination had relative standard deviations below 0.6% for both LICs used in the experiments. In general, the LIC containing isooctane showed a greater stability in measurement results and had lower leakage currents than the LIC containing tetramethylsilane, as seen in Sec. IV. This behavior was also noted in the investigation by Andersson and Tölli.¹⁴ The somewhat lesser stability of the LIC containing tetramethylsilane need not be representative for the liquid, it may also be related to the specific microLion chamber. The stability of both LICs was somewhat poorer at the polarizing voltage of 25 V as compared to the other, higher voltages, as seen in Fig. 4. However, 25 V is well below the voltage usually employed in these types of chambers. The relative standard deviations for the corrected LIC to

NACP-02 ratios for the polarizing voltages employed in the present work were all below 0.4%. Beyond the relative standard deviations of the general collection efficiencies, given by the spread in calculation results from the different combinations of measurement results from various dose rates (d_i) between 0.2 and 1.0 Gy min⁻¹, there are other possible sources of uncertainty to consider. The initial activity of ¹⁸F, and therefore also the dose rates (d_i) at which the general collection efficiencies were determined, had a relative standard deviation of 0.2% for the different experiments after correlation to the lowest cyclotron yield. Uncertainties in the general collection efficiencies may also come from small variations in the relative positioning of the radiation source and the ionization chambers, as previously discussed. These uncertainties are not easily quantified, but should be very minor given that the LIC and NACP-02 chambers were fixed in a custom perspex holder and that the different measurement series were correlated to the batch with the lowest cyclotron yield. This gives a consistency in the relationship between dose rate in the LIC and NACP-02 chambers, which in turn gives reliability in the ion chamber readings that are used in the two-dose-rate method. The perturbations observed will have influenced the measurement results of the leakage currents. Since the leakage currents were subtracted from the measurement results for the different LIC polarizing voltages, the perturbations have thus affected the calculation results for the general collection efficiencies. As noted in Sec. IV, the maximum leakage currents measured amounts to 0.4 and 1.0% of the ionization currents at the lowest dose rates and polarizing voltages used in the determination of the general collection efficiencies for isooctane and tetramethylsilane, respectively. The uncertainty in the measured leakage currents thus lends a small uncertainty to the calculated general collection efficiencies at the lowest dose rates. Negative effects from uncertain leakage currents at higher dose rates can be considered as negligible. However, it cannot be ruled out that the perturbations, which led to the exclusion of measurement results below an estimated dose rate of 0.2 Gy min⁻¹, were not present also at higher dose rates for the LIC polarizing voltages used although in a very limited capacity. The manufacturer recommends a polarizing voltage of 800 V (range 400 - 1000 V) for the microLion chamber. Lower voltages were employed in this investigation, as the general recombination effects studied were found to be negligible over 300 V for the dose rates used in this work. Another possible uncertainty for the results in the present work is that the polarity effect has not been taken into account

for any of the ionization chambers used. The polarity effect, which is specified to $< 1\%$ by the manufacturer of the microLion chamber, was investigated by Chung *et al.*⁹ in a work that evaluated the chamber performance in radiotherapy reference dosimetry of nonstandard fields. The authors found that the ratio of polarity correction for an IMRT to a reference ($10 \times 10 \text{ cm}^2$) field was in the range $0.998 - 1.004$, with a measurement uncertainty of $0.02 - 0.14\%$. Corrections of this magnitude can be considered smaller than the other uncertainties discussed for the present work.

As the two-dose-rate method used in this work has been derived from the Greening theory¹⁵ it is prudent to assume that it has inherited limiting factors from the underlying theory. Greening found that the theory was valid for air-filled ionization chambers for collection efficiencies above 70% . The Greening theory, which is a simplification of the more general theory by Mie,²² has not been theoretically verified for liquids. However, there have been several efforts that employ the Greening theory, either directly or as a part of experimental methods, with good results for LICs.^{10,12,14} Pardo-Montero and Gómez¹² have presented a three-voltage method for continuous beams, which is based on the Greening theory for general recombination and also the theory by Onsager²¹ for initial recombination. The authors reported a good agreement between the Greening theory and results, from experiments performed in continuous beams from a ^{60}Co unit, for a modified two-voltage method and the three-voltage method for continuous beams for general collection efficiencies above 96% . Furthermore, the Greening theory was observed to be valid for LICs for general collection efficiencies above 90% , in a comparison with an experimental method using 140 keV photons by Johansson and Wickman.¹⁰ Andersson and Tölli¹⁴ found that the two-dose-rate method, based on the Greening theory, achieved good results for 120 kVp x-ray photons over approximately the same interval of general collection efficiencies. In the present work, which employs 511 keV annihilation photons, the corrected LIC to NACP-02 ratios indicate that the two-dose-rate method achieves good results in signal to dose linearity for a larger interval of general collection efficiencies ($0.7 < f < 1$). A comparison between the experimental and the theoretical collection efficiencies in the present work also support this conclusion. However, there are deviations up to 8% for the LIC containing iso-octane operated at 25 V . The largest deviation here is found for the highest dose rate, where the theoretical

collection efficiency was calculated to 0.685. This is outside the valid region of collection efficiencies according to the Greening theory that was used to derive the present two-dose-rate method. As noted in Sec. IV, there are also smaller deviations between experimental and theoretical results for the LIC containing tetramethylsilane, but here no systematic relation to dose rate or polarizing voltage can be found. This is likely related to the somewhat poorer stability of the LIC containing tetramethylsilane. A broader range of applicability in valid general collection efficiencies for the two-dose-rate method is naturally beneficial as it allows for viable corrections for the general recombination effect at higher dose rates. The present work does, however, indicate that the valid range of collection efficiencies is dependent on photon energy. The energy dependence of LICs, and more specifically the observed variation in valid range of collection efficiencies for the present two-dose-rate method may be tied to the initial recombination effect. The possible connection between initial recombination and energy dependence for LICs is that the radiation quality and type will determine the density of ions created along an incident ionizing particle track. As previously discussed, the two-dose-rate method for continuous beams does not explicitly model initial recombination and furthermore, the underlying theory by Greening may not be valid for liquids in all relevant aspects. The experimental findings in the present work thus suggest that further studies on the underlying theories for the initial and general recombination effect in liquids are needed if LICs are to become a viable option in high precision radiation dosimetry.

The experimentally determined general collection efficiencies for comparable polarizing voltages and dose rates are similar for the liquids used in the present work, excepting results from 25 V polarizing voltage. In contrast, for the same liquids used in measurement of 120 kVp x-ray photons, Andersson and Tölli¹⁴ found differences between the general collection efficiencies, and in particular the corrected measurement results for isooctane and tetramethylsilane. The ratio between the corrected measurement results for tetramethylsilane and isooctane for all dose rates and LIC polarizing voltages was 1.5 (relative standard deviation 1.5%) in the present work. This can be compared to the corresponding ratio, which was 5.9 (relative standard deviation 1.7%), for the results from measurements of 120 kVp x-ray photons. These ratios display a small systematic increase with

increasing LIC polarizing voltage for both photon energies, except for the highest voltage in the different experiments where the corrections for general recombination losses according to the two-dose-rate method were very close to unity. The deviations in the results for the different photon energies may be explained by the findings of Wickman *et al.*¹⁶ where mass-energy absorption coefficients were employed to discuss a reported energy dependence of isooctane and tetramethylsilane employed as sensitive media in LICs. In Fig. 6 the general collection efficiencies, as a function of the corrected ionization currents (I), from Andersson and Tölle¹⁴ are compared to the present work for isooctane and tetramethylsilane.

FIG. 6. A comparison of experimentally determined general collection efficiencies according to the two-dose-rate method for 120 kVp x-ray (∇), and 511 keV annihilation photons ($\circ$). The collection efficiencies are displayed as a function of the ionization current corrected for general recombination, i.e. the ionization current escaping initial recombination.

The results from the measurements of 120 kVp x-ray photons were extrapolated (second order polynomial) to make a comparison to the results in the present work, as seen in Fig. 6. The extrapolations are based on the results from 120 kVp x-ray photons since the experimental uncertainties were very small in these experiments. For the LIC containing isooctane operated at 100

V in the measurements of 120 kVp x-ray photons, there are a few general collection efficiencies that are slightly below 90%, and this lends a small uncertainty to the extrapolation. For the case of the LIC containing tetramethylsilane operated at 100 V there was only a single general collection efficiency above 90% in the results from the experiments involving 120 kVp x-ray photons (i.e. the lowest corrected ionization current), and this comparison can thus be viewed as more uncertain than the other three cases. From the extrapolations there is an agreement within 0.7% for both isooctane and tetramethylsilane, which is considered to be within the experimental uncertainties for the present work, as previously discussed. To compare the relative agreement in the comparisons, the absolute differences between the extrapolations and the results in the present work were analyzed. The best agreement was found for isooctane operated at 100 V and the worst was found for tetramethylsilane operated at 100 V. This may be explained by that the general collection efficiency was only above 90% for the lowest dose rate in the 120 kVp x-ray photon results for the LIC containing tetramethylsilane operated at 100 V. The two-dose-rate method has thus been found to correct LIC measurement results with respect to the general recombination effect, so that the resulting ionization currents are comparable at the two photon energies considered here.

VI. CONCLUSIONS

The two-dose-rate method for continuous beams applied to measurements of 511 keV annihilation photons from the radioactive isotope ^{18}F gives a good correlation, in signal to dose linearity, between a LIC and a reference ionization chamber for general collection efficiencies above 70%. The method leads to corrections for the general recombination effect, which gives comparable corrected ionization currents in the low- to medium photon energy regime. However, a difference has been found in the valid range of general collection efficiencies when comparing the present results with a previous work involving 120 kVp x-ray photons. Measurements by a monitor ionization chamber can, in the two-dose-rate method for continuous beams, be substituted by an analytical expression involving exponential decay when performing measurements on radioactive isotopes. This may be beneficial when corrections for general recombination losses are needed in experimental setups where monitor chambers are difficult to incorporate. The microLion chamber has been found to be sensitive to

ambient electric fields, resulting in perturbations in the measurement signal. The usability of LICs is limited at very high dose rates in continuous beams, where there are presently no methods that can be used to correct for general recombination losses. At low dose rates, LICs may have problems with leakage currents, and specifically for the microLion chamber there may be additional problems with perturbations caused by ambient electric fields. Increasing the chamber polarizing voltage, which diminishes the general recombination effect, has been found to make the microLion chamber more sensitive to ambient electric fields. Prudence is thus advised when employing the microLion chamber in radiation dosimetry, as ambient electric fields of the strength observed in the present work can be found in many common situations. Due to the uncertain theoretical basis for recombination losses in liquids, further studies on the underlying theories for the initial and general recombination effect are needed if LICs are to become a viable option in high precision radiation dosimetry.

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