Reducing the background of secondary ions in an ion-counting nanodosimeter

G. Hilgers¹ and H. Rabus

Physikalisch-Technische Bundesanstalt,
Bundesallee 100, 38116 Braunschweig, Germany

E-mail: gerhard.hilgers@ptb.de

Abstract: In previous investigations using the PTB Ion Counter nanodosimeter, significant deviations for large cluster sizes were found in the comparison between measured and simulated data of ionisation cluster size distributions. These deviations could be explained quantitatively using a simple parametric model for a background of secondary ions, which are produced within the transport system of the target gas ions to the ion detector. To correct for these secondary ions, unfolding procedures were applied to the measured data to remove the effects of this background. However, the parameters of the model describing the background of secondary ions strongly depend on operational parameters of the nanodosimeter, and any change in these operational parameters requires a new characterisation of the model’s parameters. In this work, the influence of the nanodosimeter’s operational parameters on the background of secondary ions and the collection efficiency of the detected ions was therefore systematically investigated. At the end of this process operational conditions were found where the background of secondary ions is reduced substantially while the effect on the spatial distribution of the collection efficiency is still acceptable. For some beam geometries of particular relevance for radiation biology, cluster size distributions measured with these new operational parameters can be used directly so that the unfolding procedures, and consequently the characterisation of the secondary ion background with respect to the parameters required by the corresponding model used in the unfolding, can be omitted.

Keywords: Interaction of radiation with matter; Microdosimetry and nanodosimetry

¹Corresponding author.
1 Introduction

The pattern of inelastic interactions in subcellular targets, especially in DNA, is substantially responsible for radiation-induced damage to tissue [1, 2]. Therefore, the track structure of ionising particles is of particular importance in view of the biological effectiveness of ionising radiation [3, 4].

Nanodosimeters are radiation detectors measuring the relative frequency distribution of the ionisation cluster size, which is characteristic for the experimentally accessible ionisation component of the particle track structure [5]–[7]. The ionisation cluster size denotes the number $\nu$ of ionisations created in a target volume by a primary particle and all its secondaries. A primary particle can either traverse the target volume or pass it at a distance $d$ with respect to its centre (figure 1). The ionisation cluster size produced in the target is a stochastic quantity that results from the superposition of the pattern of ionisations within a particle track and the geometrical characteristics of the target volume. The ionisation cluster size distribution represents the statistical distribution of the probabilities $P_{\nu}(Q,d)$ that exactly $\nu$ ions are created in the target volume for radiation quality $Q$ with the primary particles passing the target volume at a distance $d$. Often the mean ionisation cluster size $M_1(Q,d)$ is of particular interest, which is defined by:

$$M_1(Q,d) = \sum_{\nu=0}^{\infty} \nu \cdot P_{\nu}(Q,d).$$

In previous investigations with the PTB Ion Counter nanodosimeter, the comparison of measured and simulated data of ionisation cluster size distributions showed substantial deviations in the frequency of occurrence for large cluster sizes [8, 9]. These deviations could be explained by a background consisting of additional ionisations, which are produced within the transport system for
Figure 1. Schematic representation of the creation of an ionisation cluster by an ionising particle passing by a cylindrical target volume of diameter $D$ at a distance $d$ from the cylinder axis. In the shown segment of the particle track, the solid circles represent the locations of ionisation interactions.

the ionised target gas molecules. In order to determine the “true” cluster size distributions or derived quantities from the measurements, a model describing the background of secondary ionisations was developed for use in unfolding procedures, which were applied successfully to the measured data to remove the effects of the background due to secondary ionisations [10]. However it was found, that the parameters of the model describing the background of secondary ions strongly depend on those operational parameters of the nanodosimeter which are related to the target gas (i.e. type of target gas and target gas pressure) and to the drift time window width. A change in any of these operational parameters requires a new characterisation of the parameters of the model describing the secondary ion background, which does not seem feasible in practice. Other operational parameters, which are not related to the target gas, were kept constant in these measurements and therefore did not enter the model explicitly. However, implicitly the influence of these parameters is contained in the parameters describing the model. Since the additional ions are produced within the ion optics, it is to be expected that the operational parameters related to the ion optics will affect the secondary ion background as well. Therefore, these operational parameters, i.e. the operational voltages of the ion optics’ electrodes and the drift voltage, were systematically modified with the objective to reduce the background of secondary ions to a negligible amount — or at least as much as possible — while simultaneously preserving the collection efficiency at a maximum possible level, in order to obtain track structure information that is as undistorted as possible.

2 Setup of the experiment

2.1 PTB ion counter

Ionisation cluster size distributions can be measured with an ion-counting nanodosimeter. The original setup of the device is described in [11]; later improvements and an improved characterisation are described in [9]. The nanodosimeter shown schematically in figure 2 consists of an interaction
region filled with a rarefied target gas, an electrode system to extract ions from the interaction region, an evacuated acceleration stage with an ion-counting detector at its end, and a primary particle detector.

The interaction region of the nanodosimeter is within the electrodes of a plane parallel plate capacitor and is filled with the target gas at a pressure in the order of 1 mbar. A primary ion traversing the interaction volume ionises the target gas molecules, and after leaving the interaction volume it is registered in a semiconductor detector, which triggers the data acquisition. Due to the electric field, which is applied across the plane parallel plate capacitor, the ionised target gas molecules produced by the primary ion and its secondaries drift towards the lower electrode. Ions produced in the target volume, which is located inside the interaction region right above a small aperture in the lower electrode, are extracted from the interaction volume through this extraction aperture. After extraction, the ions are transported and accelerated in an ion optics to an ion counting secondary electron multiplier, where they are then detected. The shape and the size of the target volume are defined by the spatial distribution of the collection efficiency of the generated ions, which is primarily determined by the electrical field strength in the interaction volume as well as the working gas and its pressure. Repeating this measurement for a large number of single primary particles of radiation quality \( Q \) and distance \( d \) yields the relative frequency distribution \( P_\nu(Q, d) \) of the ionisation cluster size \( \nu \) of detected ions.

The measured ionisation cluster size distributions contain a background which arises from secondary ions produced due to the scattering of extracted target gas ions within the ion transport.
Figure 3. Sketch of the ion transport optics of the ion-counting nanodosimeter. The target gas ions created in the interaction chamber are extracted through the extraction aperture, transported and accelerated in an ion optics towards an ion detector. ⊕ denotes target gas ions scattered within the ion optics (here exemplarily scattered onto an electrode); ⊖ denotes secondary electrons created by the scattered target gas ion, which are accelerated towards the extraction aperture.

In ion transport simulations, scattered target gas ions were found to impinge on the surface of the electrodes of the ion transport system, with kinetic energies of up to 2.5 keV. When impinging on a surface, ions of this energy are able to liberate secondary electrons. Additionally, secondary electrons can be created in the collisions of the target gas ions with the neutral target gas molecules expanding through the extraction aperture. These secondary electrons are accelerated towards the extraction aperture, gaining an energy of up to 2.5 keV in the region close to the extraction aperture, which is on ground potential. Due to the target gas expanding out of the interaction volume, the gas density is at its greatest immediately downstream of the extraction aperture. Therefore, these secondary electrons are able to create additional ions from the neutral target gas molecules expanding through the extraction aperture. These secondary electrons add to the ionised gas molecules originating from ionisations due to the primary particle. Since the contribution from primary particles cannot be distinguished from the contribution of secondary ions, the size of the measured ionisation cluster increases.

2.2 Position-sensitive detection of the primary particle

To allow for the imaging of the spatial distributions of the collection efficiency, the trigger detector was replaced by a two-dimensional position-sensitive detector (PSD).

The active area of the position-sensitive detector used for measurements is 20 mm in height and 20 mm in width (First Sensor, DL400-7 [12]). The PSD is not pixel-based, but covered with a resistive layer on each side, the top and the bottom of the silicon. The two layers are contacted orthogonally with respect to each other, one along the “height axis” and the other one along the “width axis”. Both resistive layers work according to the charge division principle. The impact
position of the primary particle with respect to the centre of the detector is calculated according to:

\[
X = L_X \cdot \frac{(Q_{X1} - Q_{X2})}{(2 \cdot (Q_{X1} + Q_{X2}))}
\]
\[
Y = L_Y \cdot \frac{(Q_{Y1} - Q_{Y2})}{(2 \cdot (Q_{Y1} + Q_{Y2}))}
\]

Here \(X\) and \(Y\) are the distances from the detector centre, \(L_X\) and \(L_Y\) are the lengths of the detector in the x- and y-direction, and \(Q_{X1}\) and \(Q_{X2}\), and \(Q_{Y1}\) and \(Q_{Y2}\), are the charges collected at the two detector output terminals in the x- and y-direction, respectively. The sum of both charges in each direction represents the energy of the primary particle.

Although the active area of the PSD is not divided into well-defined separate pixels but is a "continuous" detection area, in the data processing virtual rectangular pixels of arbitrary size can be configured independently for each direction.

The electronic configuration, the signal read-out and the signal processing of the two-dimensional PSD is carried out in analogy to the one-dimensional PSD as described in [8]. Moreover, the steps taken in [8] to prevent distortions of the waveform of the resistive-feedback charge-sensitive preamplifiers due to charge equalisation by the RC-networks connecting the PSD to the charge-sensitive preamplifiers were applied here as well.

2.3 Characterisation of the position-sensitive detector

The uncertainties encountered in the experiment are described in detail in [9]. Due to the upgrade of the nanodosimeter with the two-dimensional PSD, four additional contributions have to be taken into account. These parameters are the position resolution of the PSD and the linearity of the determination of the position of the particle impinging on the PSD’s surface, both for the x-axis and the y-axis. In order to determine these four parameters, two crossed grids were placed in front
Figure 5. Difference image between the measured and the reconstructed “digital” intensity profile. The colour represents the state of the respective pixel: (i) grey ("0") means no difference between measurement and reconstruction, i.e. measurement and reconstruction coincide, (ii) black ("1") corresponds to slits ("1") in the measurement and strips ("0") in the reconstruction, and (iii) white ("-1") corresponds to strips ("0") in the measurement and slits ("1") in the reconstruction.

of the detector close to the detector’s surface, with both a slit width and a strip width of 1 mm. Then the detector was irradiated with alpha particles from a $^{241}$Am source. Figure 4 shows the intensity distribution of alpha particle hits impinging on the detector surface as a function of the x-coordinate $d$ and the y-coordinate $h$ on the PSD with $d = h = 0$ mm representing the centre of the PSD. The width and the height of the virtual pixels is $50 \, \mu m \times 50 \, \mu m$. The slits and the strips of the crossed grids in front of the detector are clearly visible. In the centre of the PSD, the crossed grids are imaged almost distortion-free, whereas towards the corners of the detector the degree of distortion increases significantly. Specifically, it can be seen that the image in fact is mainly affected by distortions in the areas close to the corners, but in the areas in the centre of the detector edges it is also almost as distortion-free as in the central area of the PSD. However, it was found that the image of the crossed grids had shrunk by a factor of 0.96 in the x-direction and by 0.91 in the y-direction.

The position resolution is defined as the spacing between the two points along a line across a step-like intensity profile where 10% and 90% of the maximum intensity are measured. Numerically, the intensity profile can be approximated by the convolution of a rectangular distribution representing the grid and a Gaussian distribution representing the smearing of the intensity profile across the sharp edge. Then, the position resolution is identical to $2.563 \sigma$ of the standard deviation of the Gaussian distribution (or $1.088 \sigma$ of its FWHM).

In order to obtain the overall position resolution in each direction over the whole length of the detector surface, the number of pixels with the number of counts between 10% and 90% of the mean value of the number of counts of the pixels in the plateau corresponding to the slits of the crossed grids was determined and averaged over the whole intensity profile for both directions independently.
However, only those data inside a limited rectangular area of $-8.6 \, \text{mm} \leq d \leq 8.25 \, \text{mm}$ and $-7.85 \, \text{mm} \leq h \leq 8.3 \, \text{mm}$ centred with respect to the detector surface were taken into account in order to omit those data points at the corners of the detector where the image was distorted.

The relative root mean square (r.m.s) detector non-linearity $\delta_d$ ($\delta_h$) on the x-axis (y-axis) for the position is determined as [13]:

$$
\delta_d = \frac{\sqrt{\langle (d_m - d_t)^2 \rangle}}{L_x}
$$

$$
\delta_h = \frac{\sqrt{\langle (h_m - h_t)^2 \rangle}}{L_y}
$$

with $d_m$ ($h_m$) and $d_t$ ($h_t$) corresponding to the measured and the true coordinates of the edges of the slits of the grids, respectively, and $L_x$ ($L_y$) denoting the active length of the PSD in the x-direction (y-direction). Since the position of the grids with respect to the detector surface was not precisely reproducible, the exact position of the grids relative to the detector was reconstructed from the measured intensity profile. Figure 5 shows the difference image between the measured and the reconstructed “digital” intensity profile. The colour represents the state of the respective pixel: (i) grey (“0”) means no difference between measurement and reconstruction, i.e. measurement and reconstruction coincide, (ii) black (“1”) corresponds to slits (“1”) in the measurement and strips (“0”) in the reconstruction, and (iii) white (“−1”) corresponds to strips (“0”) in the measurement and slits (“1”) in the reconstruction. In the data derived from the measured intensity profile the intensity is set to “1” if the number of the counts in the pixel is larger than half of the mean value averaged over all pixels in the considered section of the PSD’s active area and is set to “0” otherwise. Setting the threshold to half of the mean value of the counts in the pixels is motivated by the ratio of the strip area to the slit area of the crossed grids of 3:1 with the border lines of the section under consideration located in the middle between the rows and columns of the slits. From the resulting (digital) intensity profile for both directions, the positions of the edges of the slits of the crossed grids are determined. To obtain the reconstruction data set, an intensity profile of crossed grids with a slit width and a strip width of 1 mm each was shifted such that the positions of the edges of the slits of the crossed grids coincide best with the measured intensity profile. The detector non-linearity was then calculated from these two data sets.

For the section of the active area between $-8.6 \, \text{mm} \leq d \leq 8.25 \, \text{mm}$ and $-7.85 \, \text{mm} \leq h \leq 8.3 \, \text{mm}$ (dashed line in figure 5) the position resolution in the x-direction was determined to be 320 $\mu\text{m}$ and in the y-direction 370 $\mu\text{m}$, leading to corresponding Gaussian distributions of $\sigma = 125 \, \mu\text{m}$ for the x-direction and $\sigma = 144 \, \mu\text{m}$ for the y-direction. Together with the detector non-linearity of 100 $\mu\text{m}$ in the x-direction and 60 $\mu\text{m}$ in the y-direction the total standard uncertainty in the determination of the position where the primary particle hits the detector amounts to $\pm 160 \, \mu\text{m}$ in the x-direction and $\pm 156 \, \mu\text{m}$ in the y-direction. Since the spatial distribution of the collection efficiency for the ionised target molecules is shaped such, that its image covers only the section on the PSD surface with $-5 \, \text{mm} \lesssim d \lesssim 5 \, \text{mm}$ (see figure 7), it is justified to take into account only this section. Therefore, for the section between $-4.9 \, \text{mm} \leq d \leq 4.8 \, \text{mm}$ and $-7.85 \, \text{mm} \leq h \leq 8.3 \, \text{mm}$ (solid line in figure 5) the position resolution in the x-direction was 310 $\mu\text{m}$ and in the y-direction 350 $\mu\text{m}$, with corresponding Gaussians of $\sigma = 121 \, \mu\text{m}$ for the
x-direction and \( \sigma = 137 \, \mu m \) for the y-direction. Together with the detector non-linearity of 35 \( \mu m \) in the x-direction and 40 \( \mu m \) in the y-direction the total uncertainty in the determination of the position where the primary particle hits the detector amounts to \( \pm 126 \, \mu m \) in the x-direction and \( \pm 143 \, \mu m \) in the y-direction.

3 Material and methods

As mentioned earlier, changing the operational voltages of the ion optics apertures and the drift voltage not only influences the processes that lead to excess ionisations due to collisions of ions within the extraction ion optics, but will also change the collection efficiency in the target volume. Knowledge of the spatial distribution of the collection efficiency is essential for allowing simulations to be compared with measured ionisation cluster size distributions.

In the measurements in this work, the ionisation cluster size was therefore recorded together with the impact position \((d,h)\) of the primary particle on the PSD, where the particle hit also triggers the data acquisition. The position where the primary ion hits the PSD defines the end point of its trajectory. Together with the known location of the ion source, this allows the primary ion’s trajectory to be reconstructed. Hence, the measured ionisation cluster size distributions for different impact positions yield information about the spatial distribution of the collection efficiency. More precisely, plotting the mean ionisation cluster size \(M_1(d, h)\) as a function of \(d\) and \(h\) results in an image of the projection of the spatial distribution of the collection efficiency along the primary ion’s trajectory and thus of the projection of the target volume. Since the collection efficiency is of cylindrical symmetry with respect to the central axis of the extraction aperture, \(M_1(d, h)\) represents the Abel-transformed spatial distribution of the collection efficiency onto the surface of the PSD. If the coordinate system is defined such that the y-axis on the detector is within the symmetry plane of this projection, \(d\) refers, in terms of detector coordinates, to the distance between the primary ion trajectory and the central axis of the extraction aperture. The x-axis can then be chosen such that it passes through the centre of the PSD surface.

In order to identify optimum operational parameters of the nanodosimeter for a reduced background of secondary ions, measurements of ionisation cluster formation in 1.2 mbar \(C_3H_8\) and in 1.2 mbar \(H_2O\) were carried out using as primary ions alpha particles from a \(^{241}\)Am source while varying the operational voltages of the electrodes in the ion optics and the drift voltage between successive measurements. As the impact position of the primary particle was recorded together with the ionisation cluster size, the changes of the secondary ion background in the ionisation cluster size distributions and the effect of the modified voltages on the spatial distribution of the collection efficiency were monitored simultaneously. The optimum values that were finally adopted as new operational voltages are substantially lower than the original voltages published in [14]. However, the ratios of the voltages of the apertures of the ion optics and the drift voltage are preserved, thus effectively preserving the imaging properties of the ion optics.

In order to obtain a high-quality image of the collection efficiency on the PSD and to obtain good statistics in the ionisation cluster size distributions for the individual virtual pixels on the PSD, a large number of events is required. On the other hand, due to the long drift time of the target gas ions to the extraction aperture (up to 200 \(\mu s\) for \(C_3H_8\) and 250 \(\mu s\) for \(H_2O\) for the new operational voltages), only a low primary particle rate is allowed to prevent pile-ups in the data acquisition of
the target gas ions. Due to the combination of these two constraints, these measurements were not feasible at accelerator facilities because of the long time required for the measurements. Therefore, all measurements were carried out using a $^{241}$Am nuclide source.

### 3.1 Ion optics potentials

The accelerating potential $U$ along the central axis of the ion optics below the extraction aperture is shown in figure 6 for the old (HV) and the new (LV) set of operational voltages. Here $a$ denotes the distance downstream from the extraction aperture, which is at $a = 0$ mm and is on ground potential. The dotted lines indicate the positions of the electrodes of the ion optics (see also figure 3). According to the much lower potential of the new operation conditions, the kinetic energies of the ionised target gas molecules scattered within the ion optics are reduced, leading to a smaller number of electrons emitted in these collisions. Since the accelerating potential of these secondary electrons is decreased as well, the kinetic energy of the electrons colliding with neutral target molecules expanding through the extraction aperture is also reduced, and with it the number of additional target gas ions. Since the gas flow conditions are unchanged between the two operational conditions, both, the reduced number and the reduced kinetic energy of the secondary electrons lead to a reduction of the background of secondary ions, as seen in figures 10 and 11.

### 3.2 Spatial distribution of the collection efficiency

The effect of the change of the operational voltages on the spatial distribution of the collection efficiency is shown in figure 7 for 1.2 mbar C$_3$H$_8$. In the left plot of figure 7 the mean ionisation cluster size $M_1(d, h)$ is shown for the old configuration (HV) at $d < 0$ mm and for the new configuration (LV) at $d > 0$ mm. In the central region around $d \approx 0$ mm, $M_1(d, h)$ is somewhat
Figure 7. Measured $M_1(d, h)$ for 1.2 mbar C$_3$H$_8$ for the old configuration (HV) at $d < 0$ mm and the new configuration (LV) at $d > 0$ mm (left) and the difference of $M_1(d, h)$ for the two operational conditions $M_{1L}(d, h)$ and $M_{1H}(d, h)$ normalised with respect to the mean of $M_{1L}(d, h)$ and $M_{1H}(d, h)$ (right).

smaller for the new operational conditions than for the old ones. Due to the reduction of the drift voltage, which accompanies the reduction of the operational voltages of the electrodes of the ion optics, the drift velocity of the ionised target gas molecules is decreased, leading to an increase in the drift time, which is the time required for an ionised target gas molecule to drift from the point of its creation to the extraction aperture. With increasing drift time the increasing lateral diffusion of the ionised target gas molecules leads to a broadening of the spatial distribution of the collection efficiency, as can be observed for the outer regions for $d > 1$ mm. For the new operational conditions, $M_1(d, h)$ is somewhat larger for $d > 1$ mm than for the old ones.

These relations are quantified in the right plot of figure 7. Here the difference of $M_{1L}(d, h)$ and $M_{1H}(d, h)$ for the two operational conditions is shown normalised with respect to the mean of $M_{1L}(d, h)$ and $M_{1H}(d, h)$

$$\frac{M_1(d, h)}{M_{1L}(d, h) + M_{1H}(d, h)} = \frac{M_{1L}(d, h) + M_{1H}(d, h)}{2}.$$

In the central region, i.e. for small values of $d$, $M_{1L}(d, h)$ is about 10–15% smaller than $M_{1H}(d, h)$, with this region being widest close to the extraction aperture, i.e. for small values of $h$, and getting narrower with increasing $h$. In the region around $d = 2.5$ mm, $M_{1H}(d, h)$ is about 0–10% larger than $M_{1H}(d, h)$ for $h < 0$ mm, and for $h > 0$ mm $M_{1H}(d, h)$ is about 10–20% larger than $M_{1H}(d, h)$, thus clearly showing the broadening of the collection efficiency with increasing distance from the extraction aperture. For $d$ larger than 3.5–4 mm the collection efficiency drops to an extent making further comparison meaningless.

However, it has to be taken into account that the background of secondary ions for a target gas of 1.2 mbar C$_3$H$_8$ leads to an increase of up to 10% of the mean ionisation cluster size for $d = 0$ mm for the old operational conditions [9], i.e. the secondary ion background substantially contributes to the difference between $M_{1L}(d, h)$ and $M_{1H}(d, h)$ by increasing $M_{1H}(d, h)$ at least
for $d = 0$ mm, which means that the difference between $M_1^{LV}(d, h)$ and $M_1^{HV}(d, h)$ is effectively smaller, as indicated in figure 7.

4 Results

The effect of the variation of operational voltages on the secondary ion background is shown for two different target gases in figure 8 (1.2 mbar C$_3$H$_8$) and figure 9 (1.2 mbar H$_2$O). The plots in figures 8 and 9 show the ionisation cluster size distributions for different sets of operational voltages measured for two distances $d$, i.e. for particle tracks passing the target volume centrally ($d = 0$ mm) and tracks passing outside the target volume ($d = 4$ mm). For the sake of clarity, only a subset of the total number of voltage sets applied is shown in the plots. Here, “HV” denotes the original set of operational voltages for the ion optics’ electrodes and the drift voltage. The number in front of “HV” indicates the factor, by which the complete voltage set, i.e. electrode voltages and drift voltage, was multiplied for the corresponding measurement. For the new voltage set denoted “LV”, the operational voltages of the original set are reduced by a factor of 3. Since the cluster size depends much more on $d$ than on $h$, for the measurements a virtual pixel size of 0.5 mm $\times$ 0.5 mm was chosen to allow for a good spatial resolution in $d$. In order to obtain a sufficiently large number of particle tracks, the primary ions impinging on the detector in the pixels with centres between $h = \pm 1.5$ mm were merged, leading to an effective beam size on the detector surface of 0.5 mm width and 3.5 mm height centred at $h = 0$ mm and at $d$ as denoted in the respective plot.

For 1.2 mbar C$_3$H$_8$ (figure 8) the effect of the subsequent reduction of the operational voltages is clearly visible. For particle tracks passing the target volume centrally ($d = 0$ mm) the cluster size distributions decrease significantly more rapidly towards larger cluster sizes with decreasing operational voltages for cluster sizes $\nu \gtrsim 20$. For particle tracks passing the target volume far outside ($d = 4$ mm) this effect is already observed for cluster sizes $\nu \gtrsim 10$. The data measured for 1.2 mbar H$_2$O (figure 9) show an identical behaviour. Due to the smaller cluster sizes $\nu$ measured in 1.2 mbar H$_2$O, the decrease of the relative frequencies of cluster sizes is already observed for $\nu \gtrsim 7$ and $\nu \gtrsim 5$ for $d = 0$ mm and $d = 4$ mm, respectively. However, due to the smaller number of events measured for H$_2$O, the scatter in the data and the uncertainties are somewhat larger than for C$_3$H$_8$.

The improvement resulting from the change from the original voltages to the finally adopted voltages with respect to the secondary ion background is illustrated in figure 10 for 1.2 mbar C$_3$H$_8$. The plots in figure 10 show the ionisation cluster size distributions for different distances $d$ measured (EXP) with the original high voltages (HV) and the new modified lower voltages (LV) in comparison to the corresponding simulations (MC), which were carried out using the PTra Monte Carlo code [6, 15]. The plot denoted with $d = \pm 4$ mm represents a broad beam geometry, i.e. the beam is centred at $d = 0$ mm with a width of 8.5 mm.

The two upper plots in figure 10 show cluster size distributions for particle tracks passing the target volume centrally ($d = 0$ mm) or not centrally, but still inside ($d = 1$ mm). The reduction of the background of secondary ions is clearly visible. As already mentioned, for cluster sizes $\nu \gtrsim 20$, the cluster size distributions measured for the new lower voltages decrease significantly more rapidly towards larger cluster sizes than the distributions measured with the original higher voltages. Moreover, the measurements with the new voltages are much closer to the simulations
than the measurements with the original voltages. The simulations were carried out using spatial distributions of the collection efficiency calculated for the two sets of voltages. In the simulations, target gas ions are created in the interaction volume and are scored according to the collection efficiency at the point of their creation. For the simulations, the effect of the different sets of voltages is visible but comparatively small, indicating only a small effect of the two sets of voltages on the collection efficiency. However, the measurements with the new voltages show that the background of secondary ions has not vanished completely, but some background contribution remains, since for cluster sizes $\nu \gtrsim 50$ the relative frequency of occurrence tends to a constant value within the experimental uncertainties.

The plots for $d = 2$ mm, $d = 3$ mm and $d = 4$ mm show cluster size distributions for particle tracks passing outside the target volume. For these measurements, the effect of the change in the operational voltages on the background of secondary ions is clearly smaller than for the particle

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**Figure 8.** Cluster size distributions measured in 1.2 mbar C$_3$H$_8$ with different operational voltage sets for $d = 0$ mm and $d = 4$ mm.

**Figure 9.** Cluster size distributions measured in 1.2 mbar H$_2$O with different operational voltage sets for $d = 0$ mm and $d = 4$ mm.
Figure 10. Comparison of ionisation cluster size distributions in 1.2 mbar C$_3$H$_8$ for different distances $d$ measured (EXP) with the original high voltages (HV) and the new modified lower voltages (LV) and the corresponding simulations (MC).
Figure 11. Comparison of ionisation cluster size distributions in 1.2 mbar H$_2$O for different distances $d$ measured with the original high voltages (HV) and the new modified lower voltages (LV).
tracks traversing the target volume. Moreover, the reduction of the secondary ion background, when changing the voltages, gets less pronounced with increasing $d$. This indicates a remaining background contribution to the measured ionisation cluster size distribution, which increases with increasing $d$. Furthermore, the measurements for the new voltages for particle tracks passing outside the target volume agree less with the corresponding simulations than the measurements for particles passing inside the target volume. However, the differences between the simulations for the two voltage sets are still small.

The plot denoted with $d = \pm 4$ mm represents a broad beam geometry with the beam centred at $d = 0$ mm with a width of 8.5 mm due to the width of the virtual pixels. According to the larger width of the broad beam, the number of primary events is larger than for the previously shown cluster size distributions, consequently leading to better statistics of the measured distributions. Generally, the findings obtained for the cluster size distributions for the narrow beams with sizedistributions,consequentlyleadingtobetterstatisticsofthemeasureddistributions. Generally, the reduction of the secondary ion background, when changing the voltages, gets less pronounced with increasing $d$. Furthermore, the measurements for the new voltages for particle tracks passing outside the target volume agree less with the corresponding simulations than the measurements for particles passing inside the target volume. However, due to the better statistics of the broad beam’s cluster size distribution, the remaining background appearing in the shoulder at cluster sizes $50 < \nu < 65$ is more clearly visible than in the other cluster size distribution.

The results presented for 1.2 mbar C$_3$H$_8$ in figure 10 are confirmed in figure 11, which shows ionisation cluster size distributions for 1.2 mbar H$_2$O measured in the same experimental conditions as before for 1.2 mbar C$_3$H$_8$. However, no respective simulations are available. As before for 1.2 mbar C$_3$H$_8$, also for 1.2 mbar H$_2$O the reduction of the background of secondary ions is most pronounced for particle tracks passing the target volume centrally ($d = 0$ mm) or not centrally, but still inside ($d = 1$ mm). For measurements with particle tracks passing outside the target volume, i.e. for $d = 2$ mm, $d = 3$ mm and $d = 4$ mm, the effect of the change in the operational voltages on the secondary ion background is clearly smaller than for the particle tracks traversing the target volume. Also, the reduction of the secondary ion background, when changing the voltages, gets less pronounced with increasing $d$, and the measurement for the broad beam geometry shows a reduction of the secondary ion background similar in magnitude as for the measurements for $d = 0$ mm and $d = 1$ mm, as was also seen for 1.2 mbar C$_3$H$_8$. However, in contrast to the measurements in C$_3$H$_8$ no indications of a remaining secondary ion background are found for the measurements in H$_2$O.

This might be due to the smaller number of events measured for H$_2$O as compared to C$_3$H$_8$.

The ionisation cluster size distributions yield a mean ionisation cluster size for C$_3$H$_8$ of $M_1(d, h) = 9.2$ and for H$_2$O of $M_1(d, h) = 2.2$ for $d = h = 0$ mm, giving a ratio of $\approx 4.2$. To compare of the values of $M_1(d, h)$ for the two target gases, the scaling procedure described in [16] was applied, which has been verified experimentally in [17]. In [16] a ratio $\frac{\rho_{\lambda_{\text{ion}}}C_3H_8}{\rho_{\lambda_{\text{ion}}}H_2O} = 0.802$ is stated with $\lambda_{\text{ion}}$ being the ionisation mean free path calculated from the ionisation cross sections for C$_3$H$_8$ and H$_2$O. However, in the most recent version of PTRu the cross sections for ionisation of C$_3$H$_8$ are increased by a factor of 1.16 with respect to previous versions [15]. Not only the cross sections for protons are affected by this increase as discussed in [15], but the cross sections for all primary ions [18]. Consequently, this increase leads to a reduction of $\lambda_{\text{ion}}$ for C$_3$H$_8$. For 1.2 mbar C$_3$H$_8$ at room temperature, the mass per area is $2.17 \mu g \ cm^{-2}$. The resulting ratio $\frac{\rho_{\lambda_{\text{ion}}}C_3H_8}{\rho_{\lambda_{\text{ion}}}H_2O} = 0.69$ leads to a mass per area for H$_2$O of $3.14 \mu g \ cm^{-2}$, i.e. a target consisting of H$_2$O with mass per area of $3.14 \mu g \ cm^{-2}$ should result in the same mean ionisation cluster size as a target consisting of C$_3$H$_8$ with mass per area of $2.17 \mu g \ cm^{-2}$. For 1.2 mbar H$_2$O at room temperature, the mass per area is $0.89 \mu g \ cm^{-2}$. Therefore, a ratio of the mean ionisation cluster sizes for 1.2 mbar C$_3$H$_8$ and 1.2 mbar H$_2$O of $\approx 3.4$ is expected, taking into account that the
size of the target volume for C$_3$H$_8$ is about 4% smaller than for H$_2$O, as simulations show. Thus, the expected and the measured mean ionisation cluster sizes agree within less than 25%.

The remaining background appearing in the shoulder at cluster sizes $50 \lesssim \nu \lesssim 65$ for 1.2 mbar C$_3$H$_8$ seems not only to correlate with the distance $d$, but also with the number of ionisations produced in the target gas. Figure 12 shows cluster size distributions measured in 1.2 mbar C$_3$H$_8$ with the new lower voltages for two broad beam configurations, one with $d = \pm 1$ mm (beam width: 2.5 mm) and the other one being the sum of two broad beams with $d = -4$ mm–2 mm and $d = 2$ mm–4 mm (beam width in total: 5 mm), respectively, using the same virtual pixel size as before. For primary particles passing inside the target volume, i.e. for $d = \pm 1$ mm, where the relative frequency of ionisation clusters with cluster size $\nu \gtrsim 5$ is about one order of magnitude larger than for primary particles passing outside the target volume, also the secondary ion background is approximately one order of magnitude larger than for primaries passing outside the target volume. However, the shape of the background seems to be similar for both beam configurations.

The origin of the remaining background appearing in the shoulder at cluster sizes $50 \lesssim \nu \lesssim 65$ in 1.2 mbar C$_3$H$_8$ is unclear and will be subject of further investigation.

The ion optics’ electrode voltages and the drift voltage were not lowered below the new (LV) voltage set for two reasons. The first reason was the effect on the spatial distribution of the collection efficiency, which decreases in the central region by 10–15%, whereas in the outer regions the spatial distribution broadens, and the collection efficiency increases by 10–20%, compensating to some extent for the decrease in the centre. This amount of change is regarded as acceptable. The second reason is the drift time of the target gas ions to the extraction aperture, which increases inversely proportional with the decrease of the drift voltage. Due to the long drift time for the new (LV) voltages up to 200 $\mu$s for C$_3$H$_8$, 250 $\mu$s for H$_2$O, 500 $\mu$s for C$_4$H$_8$O (THF), only a low primary particle rate is allowed to prevent pile-ups in the data recording of the target gas ions, leading to a substantial increase in measuring time.
Table 1. New (LV) and old (HV) set of operational voltages. The voltages of the (HV) set differ from the voltages published in [14]. The voltages actually used that time were given in private communication when the device was transferred to PTB. The numbering of the electrodes corresponds to the distance from the extraction aperture: electrode 1 is closest to the aperture, electrode 4 is the most distant (see figure 3).

| Voltage set | Drift voltage | Electrode 1 | Electrode 2 | Electrode 3 | Electrode 4 |
|-------------|---------------|-------------|-------------|-------------|-------------|
| HV          | 300 V         | −286 V      | −476 V      | −810 V      | −2460 V     |
| LV          | 100 V         | −96 V       | −159 V      | −270 V      | −820 V      |

For comparison, the operational voltages for both voltage sets, the new (LV) set and the old (HV) set, are listed in table 1.

5 Summary

By changing the operational voltages of the ion optics and the drift voltage of the ion-counting nanodosimeter the background of secondary ions was substantially reduced. However, the background was not completely removed, which might in fact be impossible due to the general design principle of the nanodosimeter based on the acceleration of the target gas ions in an ion optics with neutral target gas flow inside. Comparison with simulated data show a good agreement of the measurements with the new operational parameters for ionisation cluster size distributions obtained for primary ions passing inside the target volume. For primary ions passing outside the target volume, the agreement with simulations is improved significantly due to the reduced background of secondary ions. However, the improvement is less than for primary ions passing inside the target volume. For a broad beam geometry, the agreement between simulation and measurement is similar to that for primaries passing inside the target volume. Thus, when analysing track structures of particles passing inside the target volume or track structures of a broad beam geometry, the unfolding procedures, and consequently the characterisation of the secondary ion background with respect to the parameters required by the corresponding model used in the unfolding [10], can be omitted, since the track structure information is almost undistorted by the secondary ion background. However, these beam geometries are the most relevant, since particles passing inside the target volume are most effective in creating radiation damage in relevant DNA segments, and broad beam geometries are the closest approximation to a realistic irradiation geometry.

The effect on the spatial distribution of the collection efficiency is regarded as acceptable. In the central region the collection efficiency is reduced by 10–15%, with the secondary ion background for the old operational conditions contributing substantially to the difference. In the outer regions the spatial distribution broadens, and the collection efficiency increases by 10–20%, compensating to some extent for the decrease in the centre.

Practically, the reduction of the operational voltages leads to an increase in drift time, thus reducing the maximum rate of events, which can be processed without pile-up. Therefore, measurements that require application of the two-dimensional PSD may not be feasible at accelerator facilities due to the long measuring times needed for reasonably good statistics. However, measurements at accelerator facilities can be performed using the setup described in [8], where a one-dimensional PSD is applied so that good statistics is achievable even with the new operation
voltages and the resulting longer drift times of the target gas ions. Simultaneously, on the other hand, the temporal spread of the arrival times of the ionised target gas molecules on the ion detector is widened, thus reducing counting losses due to overlapping target gas ion pulses.

All measurements published up to now [8–10, 17] were carried out using the old (HV) set of operational voltages and, consequently, are affected by the background of secondary ions. Future measurements will be carried out using the new (LV) set of voltages. Therefore, it has to be pointed out, that the ionisation cluster size distributions of future measurements will deviate from those of previous measurements in their decrease towards larger cluster sizes, which will be steeper in future measurements. Moreover, it has to be taken into account in such comparisons, that the measurements published in [8] were carried out using a target volume about half the size of the target volume used in [9, 17] and in the present investigation, due to the shorter drift time window used in [8].

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