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From micro to nanodosimetry with an avalanche-confinement TEPC: characterization with He-4 and Li-7 ions

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ABSTRACT

The measurable radiobiological effects of ionizing radiation strongly depend on the clustering of damages in subcellular sites, which are related to the particles track structure. The characteristic properties of track structure are directly measurable nowadays with bulky experimental apparatuses, which are not easily transportable and are not suited for the clinical environment. For this reason, the feasibility of new transportable detectors capable of characterizing real therapeutic beams was investigated in recent years. In particular, two novel avalanche-confinement Tissue Equivalent Proportional Counters (TEPCs) were designed and constructed for simulating nanometric sites down to 25 nm: a sealed version for operation in plain air and a new prototype with perforated walls and without external encapsulating cap for operation in a vacuum chamber with accelerated particle beams.

This work is focused on the response of the open TEPC, directly installed in the vacuum chamber of the STARTRACK nanodosimeter of the Italian Istituto Nazionale di Fisica Nucleare – Laboratori Nazionali di Legnaro (INFN-LNL), against He-4 particles from a ^{244}Cm isotopic source and 26.7 MeV Li-7 ions accelerated by the Tandem accelerator of LNL. Experimental cluster size distributions of He-4 and Li-7 ions measured with this TEPC are compared with nanodosimetric Monte Carlo simulations and with cluster size distributions measured with the STRATRACK nanodosimeter. Both comparisons highlight a very good agreement: the relative variance added by the electron multiplication process results negligible if compared to the variability of the ionization process. This encourages the use of the avalanche-confinement TEPC as a portable detector for nanodosimetric characterization of particle tracks.

Keywords: Microdosimetry; Nanodosimetry; He-4 track structure; Li-7 track structure; Tissue Equivalent Proportional Counter (TEPC); Monte Carlo simulation.

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1. Introduction

The observable radiobiological effects of ionizing radiation are strongly correlated to the clustering of damages due to ionizations in micrometer- and nanometer-sized subcellular structures, hence to the particle track structure (Conte et al., 2018). The description of the statistical fluctuations of the imparted energy in a micrometric site is achieved through microdosimetry, while the stochastics of ionization processes in a nanometric volume is the field of nanodosimetry. The characteristic properties of track structure are directly measurable nowadays with bulky experimental systems, which are not easily transportable and are not suited for the clinical environment (Bantsar et al., 2017). For this reason, an in-depth investigation about the feasibility of new transportable detectors capable of characterizing particle beams was carried out in recent years: two avalanche-confinement Tissue Equivalent Proportional Counters (AC-TEPCs), capable of simulating site sizes from the micrometric region (0.5 μm) down to few tens of nanometers (25 nm), were designed, developed and characterized. The first prototype is a sealed TEPC especially devoted to applications in plain air for measuring real hadrontherapy beams, being encapsulated in a tight vacuum aluminum shell to preserve the purity and the pressure of the internal counting gas (Mazzucconi et al, 2019; Bortot et al., 2020). The more recent avalanche-confinement TEPC has almost identical geometrical features, but it is appositively designed to be inserted in the STARTRACK apparatus of the INFN-LNL to measure the nanodosimetric properties of monoenergetic collimated particle beams, as described by Mazzucconi et al., 2020. In this experimental configuration, a direct comparison can be performed between ionization cluster size distributions measured with the STARTRACK nanodosimeter and with the TEPC simulating nanometric volumes of comparable size.

This work is focused on the response of this particular AC-TEPC against He-4 particles from a ^{244}Cm isotopic source and 26.7 MeV Li-7 ions accelerated by the INFN-LNL Tandem-Alpi accelerator. Comparison with cluster size distributions measured by the STARTRACK nanodosimeter is discussed in detail. Moreover, Monte Carlo simulations have been carried out employing a specific track structure code (Grosswendt et al., 2014) for comparison with experimental data.

2. Materials and methods

2.1. The STARTRACK nanodosimeter

STARTRACK is an experimental apparatus developed in the framework of the European research project NANDET (Radiation Quality Assessment Based on Physical Radiation Interaction at Nanometre Level). It allows to count the number of ionizations produced by single light ions within a nanometre-sized volume *V* when they cross or pass by the target volume at a specified distance (De Nardo et al., 2002). The apparatus is installed at the +50° beam line of the Tandem-Alpi accelerator complex at INFN-LNL. The main components of the track-detector are the target volume *V*, the electron collector, the drift column and a multi-step avalanche chamber (MSAC) placed on the top of the drift column. The ions of a collimated narrow beam pass the target volume *V*, a wall-less cylinder 3.7 mm in diameter and height, where they produce the ionizations that are counted by the nanodosimeter, and impinge on a solid-state detector which triggers the data acquisition.

The detector is mounted on a movable platform that allows to translate *V* orthogonally to the beam line, therefore it can directly measure the ionization events, which occur inside *V*, at different impact parameters of the primary particle trajectory with respect to the target centre. STARTRACK allows the study of the track structure properties of light ions of medical interest in order to investigate the physical quantities that correlate to the different radiobiological effectiveness of those ions (Conte et al., 2017).

2.2. The "open" avalanche-confinement TEPC

The "open" AC-TEPC was properly designed for measuring ionization distributions down to the nanometric region in the well-defined beam geometry that is available in the STARTRACK apparatus, where the detector is mounted with the main axis (anode) parallel to the direction of the collimated particle beam. This set- up allows a direct comparison between the spectra acquired by the AC-TEPC and the corresponding nanodosimetric probability distribution measured by the STARTRACK nanodosimeter. This new AC-TEPC, described in more details in Mazzucconi et al., 2020, is similar to the one described by Bortot et al., 2017, in order to make the data comparison more straightforward. It has a hollow cylindrical sensitive volume 13 mm in both diameter and height, defined by three electrodes independently biased: a graphite central anode wire

(1 mm in diameter), a cylindrical A-150 cathode shell (13 mm in internal diameter and 1 mm in thickness) and a gold-plated tungsten helix of 6 mm in diameter, made of a 100 µm wire wrapped in 19 coils.

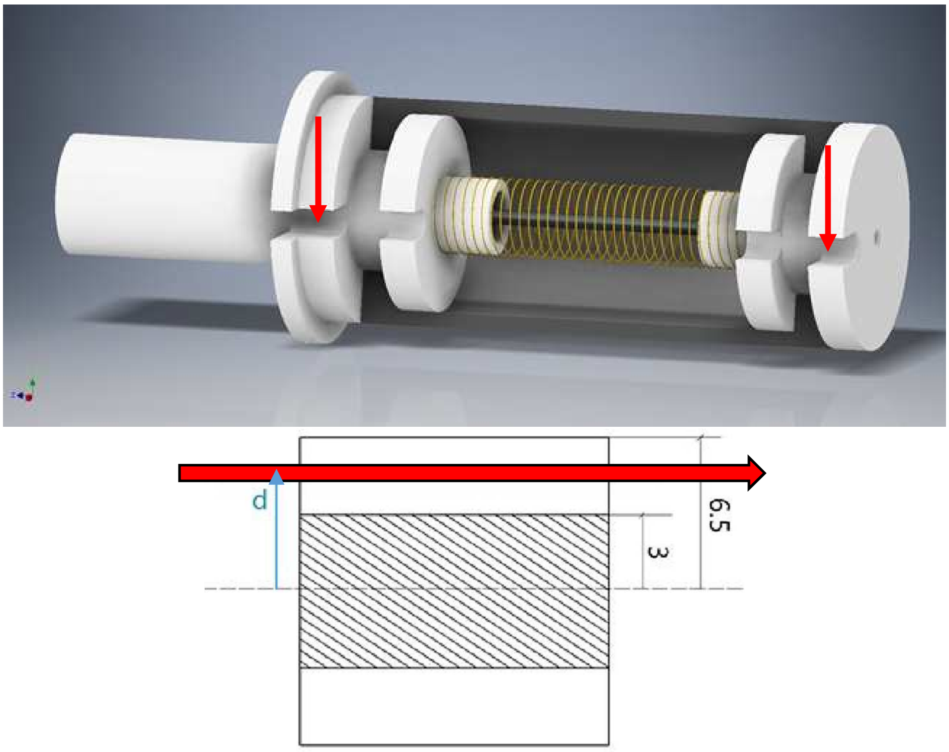

Fig.1. Top: 3D rendering of the AC-TEPC. The slits drilled in the two insulating caps for the ion beam transmission are highlighted by red arrows. Bottom: scheme of the TEPC geometry defined in the Monte Carlo code. The simulated direction of the impinging particle beam (He-4 and Li-7 ions) is indicated by a red arrow and span in the interval from 3 to 6.25 mm.

The three electrodes structure allows the separation of the TEPC gas volume into two regions: a drift region (between the helix and the cathode) and a multiplication region (between the anode and the helix). In the drift region the electrons generated by the primary particle are moved towards the helix without any multiplication, while in the multiplication region a high voltage difference is applied and generates the avalanche. By exploiting this multi-electrode structure, the TEPC can achieve a high gain without losing resolution (Bortot et al., 2017; Mazzucconi et al., 2020).

The main characteristics of this AC-TEPC consists in having a slit drilled in each of the two insulating caps (Figure 1, top), for allowing the accelerated beam to traverse first the sensitive volume of the AC-TEPC and then the sensitive volume of the STARTRACK nanodosimeter. Downstream of the two detectors, an Osram

BPX65 silicon photodiode with a square sensitive region equal to 1 mm^2 was used to trigger the data acquisition.

A customized electronic chain was developed, capable of minimizing the electronic noise (i.e. lowering the detection threshold) and allowing the acquisition of signals by both the TEPC and the silicon photodiode. The acquisition software (based on the LabView environment) allows to acquire both the voltage pulses coming from the TEPC and the photodiode. In this way different acquisition modalities can be set: without any coincidence (acquiring the microdosimeter signals only), with coincidence (microdosimeter and photodiode) and coincidence plus threshold (employing an energy threshold on the triggered events).

2.3. The irradiation set-up

The STARTRACK vacuum chamber, which was originally designed to host the whole nanodosimeter apparatus including the detector, detector handling systems and front-end electronics, was properly adapted to embed the AC-TEPC (by means of an internal optical bench) and, especially, to allow the horizontal motion of the TEPC with respect to the beam line. This experimental set-up was selected for irradiating the TEPC sensitive region at different distances *d* from the central anode wire, for characterizing the response function of the detector. The horizontal motion is controlled by a high precision nut moved by a stepper motor that can be remotely controlled. A picture of the whole experimental set-up in shown in Figure 2.

Two measurements campaigns (with 5.8 MeV He-4 particles from a ^{244}Cm source and with 26.7 MeV Li-7 ions from the Tandem accelerator of INFN-LNL) were carried out with propane gas at a density for simulating a site size of 25 nm at unit density. The calculation of the gas pressure was based on the well-known principle of simulation exploited in microdosimetry. Following this principle, for simulating 25 nm of site at unit density, the gas density should be equal to the ratio 25 nm / 13 mm, which is equal to 1.92E-6 g cm^{-3}. By considering propane and the standard conditions, the corresponding pressure is equal to 1.04 mbar. Measurements were performed at different distances *d* of the central anode from the primary particle track, in order to measure the charge collection efficiency in the drift region of the detector. For this work, the coincidence acquisition modality was adopted, recording at the TEPC only the events that at the trigger detector have an energy larger than 5.5 MeV and 25 MeV for He-4 and Li-7, respectively. These energy thresholds allow to select the events due to particles that cross the sensitive volume parallel to the anode and to discard the events produced by scattered particles.

The particle beam was collimated by means of a set of 2 aluminium collimators 0.8 mm in diameter, placed in front of the AC-TEPC and the trigger, respectively.

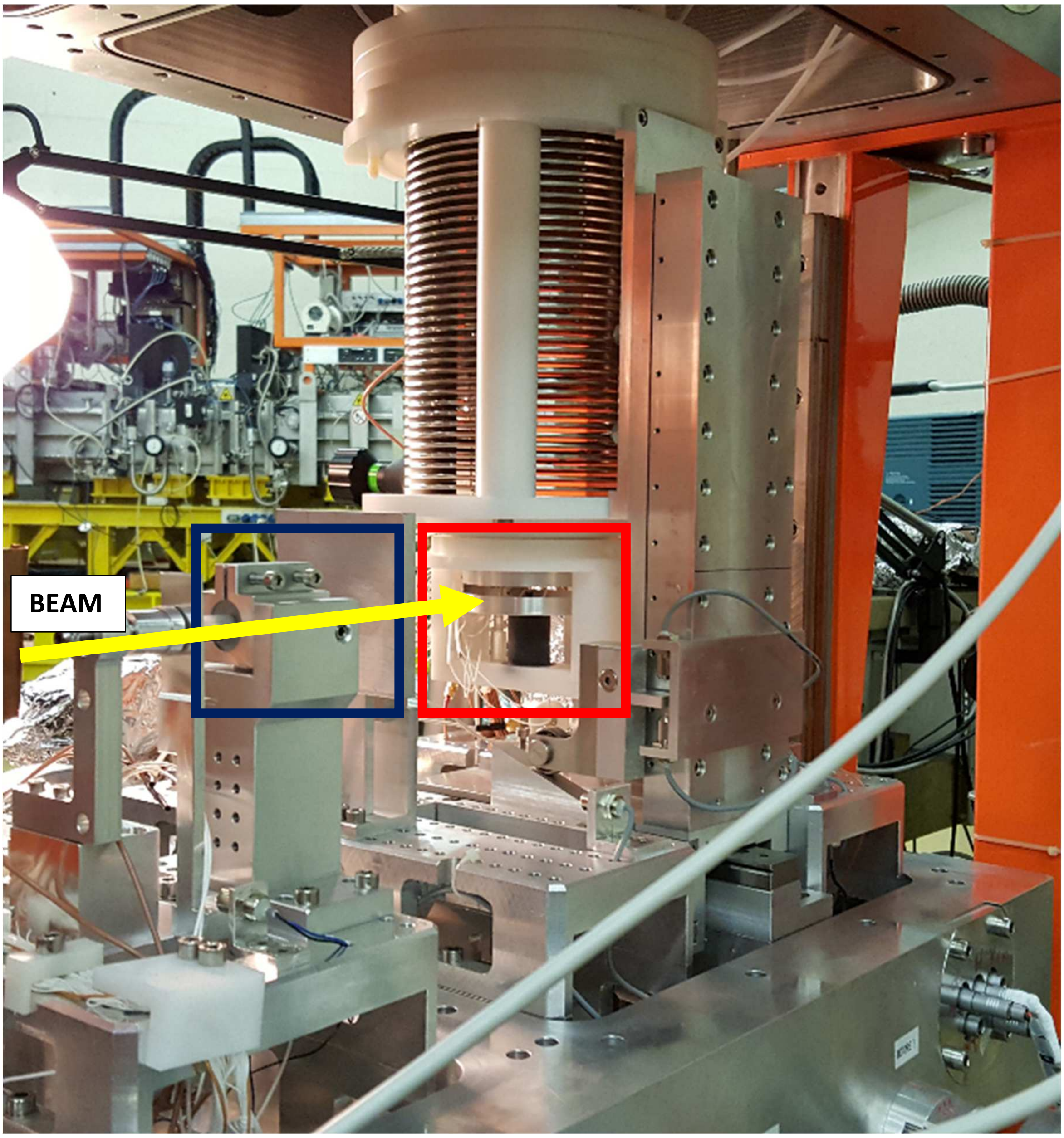

Fig. 2. Picture of the whole experimental set-up: The TEPC is embedded inside the STARTRACK vacuum chamber at the +50° beam line of the Tandem-Alpi accelerator complex at the INFN - Legnaro National Laboratories.

2.4. Monte Carlo simulations

A Monte Carlo code for track-structure simulation of light ions, originally developed by Grosswendt et al., 2014 for evaluating the cluster size distribution produced in the STARTRACK sensitive region, has been slightly modified for reproducing the sensitive region of the avalanche-confinement TEPC. Since electrons generated inside the multiplication region (between the helix and the central anode wire) undergo a smaller multiplication, their contribution to the acquired spectra has been neglected: the sensitive volume is assumed to be the drift region only (between the cylindrical cathode shell and the helix). Figure 1, bottom shows the TEPC simplified geometry implemented in the Monte Carlo code.

Several Monte Carlo simulations have been carried out for reproducing the acquired distributions for different irradiation conditions, by varying the type of particle (5.8 MeV He-4 or 26.7 MeV Li-7) and the distance *d* from the central anode wire. The beam of primary particles is directed along the detector axis with a distance *d* from 3 mm to 6.25 mm for reproducing the experimental set-up. The cluster size distribution was calculated for pure propane. In particular, a first set of simulations with He-4 particles was dedicated to the calibration of the AC-TEPC in terms of cluster size, as described in section 2.5. On the other hand, the simulations with Li-7 ions were directly compared with experimental distributions.

2.5. Data Analysis

The number ν of ions or electrons produced within *V* by a single particle, also called ionization cluster size (ICS), is measured for a number of primary particles, resulting in a frequency distribution $P(\nu)$. If the number of primary particles is large, the ionization cluster size distribution $P(\nu)$ represents the probability of measuring a number ν of ionizations in the sensitive volume *V* by a particle of a given radiation quality. In this work a total number of 10^4 events were recorded in each measurement, while simulations were performed for a total of 10^6 histories. The mean ionization cluster sizes M_1 is defined as:

$$M_1 = \sum_{\nu=0}^{\infty} \nu P(\nu) \quad (1)$$

The STARTRACK nanodosimeter has an average detection efficiency $\bar{\varepsilon}_{\mathrm{ST}} \approx 0.16$. This mean efficiency is calculated with a Monte Carlo code by simulation of the electron transport through the counter, from within the target volume to the end of the drift column. The measured mean cluster size, $M_1(\mathrm{ST})$, is lower than the initial mean number of ionizations, M_1, produced in its sensitive volume:

$$M_1(\mathrm{ST}) = 0.16 \cdot M_1 \quad (2)$$

On the other side, each initial electron released in the TEPC sensitive volume is amplified by the average gas gain factor G, therefore the mean ionization cluster size measured by the AC-TEPC, $M_1(\text{TEPC})$, is larger than the initial mean number of ionizations, M_1, produced in its sensitive volume:

$$M_1(\text{TEPC}) = G \cdot M_1 \tag{3}$$

2.5.1 AC-TEPC data elaboration

While the STARTRACK nanodosimeter directly counts the number of ionizations, ν, the output signal of the AC-TEPC is the voltage amplitude V_{out} (in mV) registered at the feedback capacitance of the charge sensitive preamplifier. To translate the voltage into a number of collected charges, an accurate measurement of the feedback capacitance is necessary. In order to convert the collected charge in terms of initial ionisation cluster size ν, the values of the gas gain *G* should also be known. In this work a different approach was used, which consists on calibrating the measured distributions of the pulse height V_{out} in terms of number of ionizations, μ, using experimental and Monte Carlo results for the He-4 ions:

$$\mu(\text{TEPC}) = (\, K \cdot V_{out}) \;; \quad K = \frac{M_1^{\text{He-4}}}{\bar{V}_{\text{out}}^{\text{He-4}}}; \tag{4}$$

Where $\bar{V}_{\text{out}}^{\text{He-4}}$ is the mean value of the measured He-4 pulse height distribution, $M_1^{\text{He-4}}$ is the mean cluster size resulting by Monte Carlo simulations. In this way the calibration factor K already includes the gas gain G. The original measured distribution $P(\mu)$ is re-binned on the subset of integer values ν, calculated as the closest integer value of μ, interpreted as the ionization cluster size.

The same calibration factor K, determined by He-4 data, is afterwards used to calibrate AC-TEPC measurements performed in the same experimental conditions (electronic chain, gas density and gas gain) with Li-7 ions.

It should be noted that this calibration coefficient holds for these specific conditions (i.e. simulated site size and electrodes voltages) for which the gas gain can be considered constant.

3. Results and Discussion

In the following sections all the distributions measured by the TEPC at 25 nm in site size will be reported and analysed, together with a comparison with the nanodosimetric simulated (through the Monte Carlo code) and experimental (from STARTRACK nanodosimeter) distributions.

3.1. TEPC cluster size distributions: comparison with Monte Carlo simulation

Figure 3 shows the cluster size distribution $P(\nu)$ of 5.8 MeV He-4 measured with the AC-TEPC at 25 nm in simulated site size, by irradiating the TEPC at a distance d = 4.75 mm (see Figure 1), i.e. in the centre of the drift region. This distribution was calibrated in terms of ν by means of the corresponding Monte Carlo simulation, by assessing the value of K in Equation 4. It should be stressed that the calibration procedure sets the mean value of the distribution only. Considered that simulations were performed in a sensitive volume of uniform efficiency of 100%, whereas in the experimental case the pulse height results as the product of the initial number of ionizations and the gas gain multiplying factor, the very good agreement in terms of both shape and width between measurements and simulations suggests a minor influence of the gas gain variance on the measured cluster size distributions.

Figure 4 shows the cluster size distribution $P(\nu)$ of 26.7 MeV Li-7 ions measured at 25 nm in simulated site size at the centre of the drift region. In this case, the measured cluster size distribution was scaled on the basis of the previous calculated calibration factor K. The same Figure shows the corresponding distribution from Monte Carlo simulation: this comparison shows that, once calibrated, the AC-TEPC is capable of assessing correctly both the mean-value and the overall shape of the cluster size distribution.

The main source of uncertainty in exploiting such a detector for the evaluation of the cluster size distribution is represented by the stability of the gas gain, which is strictly dependent on the variability of the gas pressure, the gas purity and the stability of the applied high voltages. Even if a proper and detailed uncertainty evaluation has not been performed, the gas gain has demonstrated to be stable throughout all the measurement campaigns, as highlighted by the comparison with Monte Carlo simulations for different impact parameters (Section 3.3).

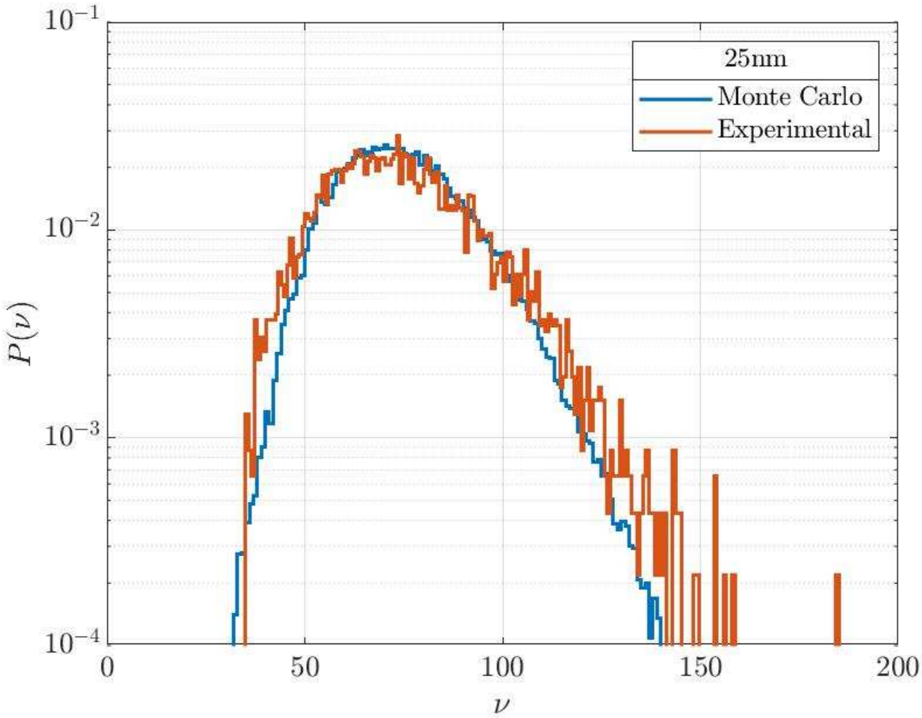

Fig. 3. Cluster size probability distributions *P(ν)* of 5.8 MeV He-4 ions at 25 nm in site size measured with the AC-TEPC (orange line) and derived from Monte Carlo simulation (blue line).

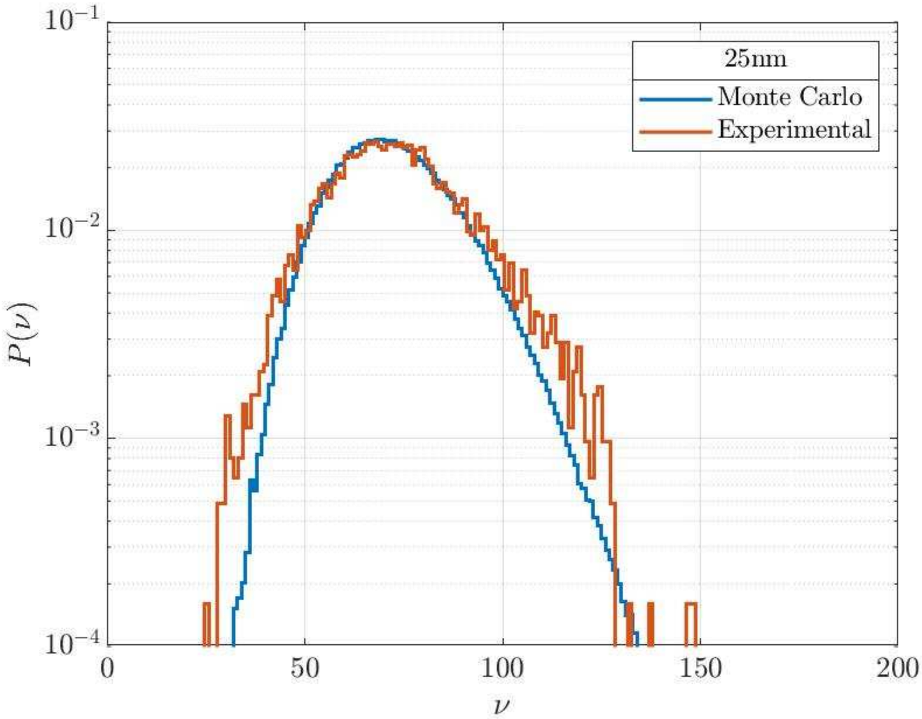

Fig. 4. Cluster size probability distributions *P(ν)* of 26.7 MeV Li-7 ions at 25 nm in site size measured with the avalanche-confinement TEPC (orange line, calibrated with the K factor, see text) and derived from Monte Carlo simulation (blue line).

3.2. TEPC cluster size distributions: comparison with STARTRACK

In order to study the possibility of exploiting the avalanche-confinement TEPC measurements to derive relevant information on the particle track structure at the nanometer level, the measured cluster size probabilities were compared with the corresponding cluster size distributions measured with the STARTRACK nanodosimeter. Whilst the site sizes of the two detectors differ only by about 20% (25 nm for the AC-TEPC and 20 nm for STARTRACK), it can be observed that the distribution measured with STARTRACK shifts to much lower cluster size values. This is due to the fact that STARTRACK nanodosimeter is characterized by a reduced detection efficiency, which decreases moving from the centre of the sensitive volume toward its border (Conte et al., 2012), with an average value defined as $\bar{\varepsilon}_{ST}$. In order to analyse correctly the shape of the two measured distributions, both the different site sizes and efficiencies of the two detectors must be taken into account.

The TEPC "reduced" distributions $P^*(\nu)$ to be compared with STARTRACK can be obtained from the original measured distributions $P(\nu)$ of Figures 3 and 4, for He-4 and Li-7, respectively, by applying the following binomial relation and taking into account the dead-time correction, as described in Conte et al., 2012:

$$P^*(\nu) = \sum_{k=\nu}^{\infty} \binom{k}{\nu} P(k)\bar{\varepsilon}^{\nu}(1-\bar{\varepsilon})^{k-\nu}$$

where $\bar{\varepsilon}$ is defined by considering the different sizes of the site (25 nm for the AC-TEPC and 20 nm for STARTRACK):

$$\bar{\varepsilon} = \frac{4}{5}\bar{\varepsilon}_{ST} = \frac{4}{5}0.16 \approx 0.13$$

Figure 5 shows the comparison between the reduced cluster size distribution $P^*(\nu)$ of 5.8 MeV He-4 measured with the AC-TEPC and the corresponding nanodosimetric spectrum measured by STARTRACK. Figure 6 shows the same comparison, but referred to 26.7 MeV Li-7 ions.

The agreement between the distributions is very good, within statistical uncertainties, and encourages the use of the avalanche confinement TEPC as a portable detector for nanodosimetric characterization of particle tracks. Moreover, it can be concluded that the relative variance added by the electron multiplication process is negligible if compared to the variability of the ionization process, at least for He-4 and Li-7 ions.

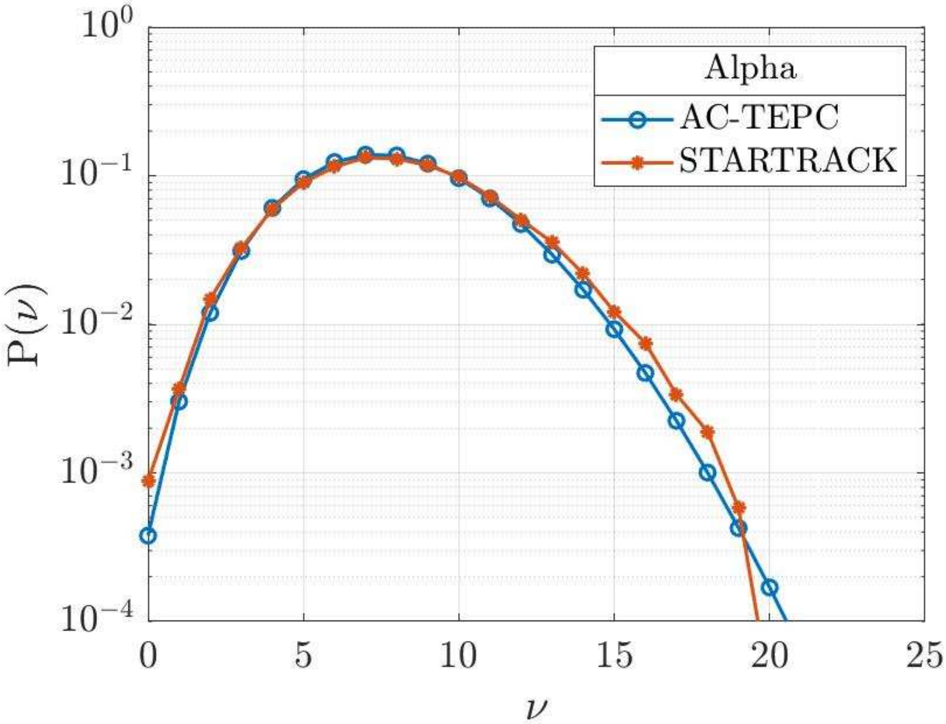

Fig. 5. Reduced cluster size probability distribution P*(v) of 5.8 MeV He-4 ions from the avalanche-confinement TEPC (blue line) and the corresponding cluster size distribution *P(v)* measured with STARTRACK (orange line).

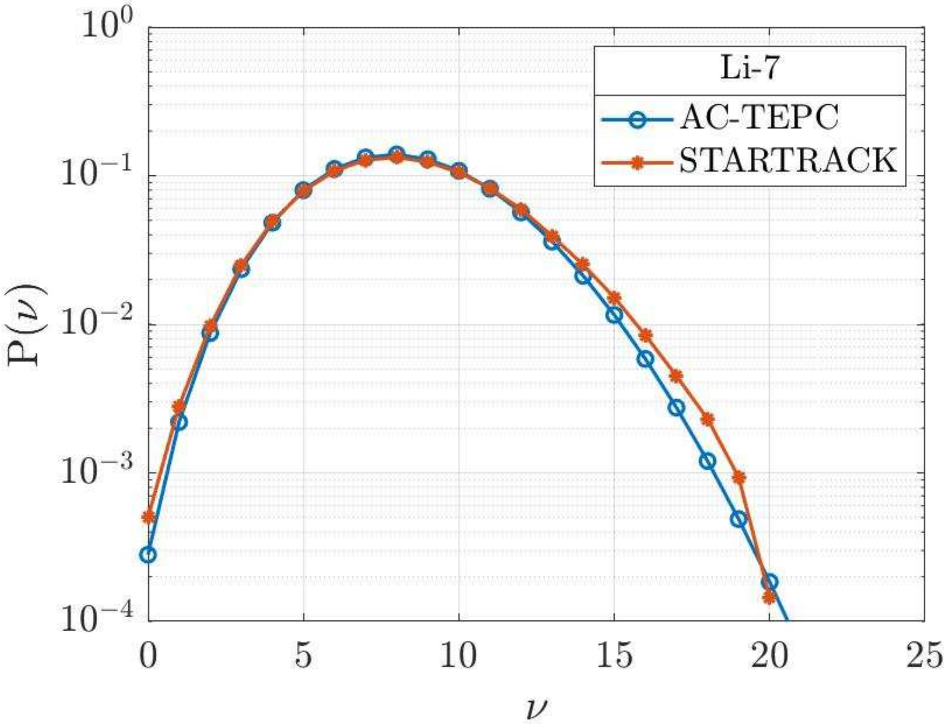

Fig. 6. Reduced cluster size probability distribution P*(ν) of 26-7 MeV Li-7 ions from the avalanche-confinement TEPC (blue line) and the corresponding cluster size distribution *P(ν)* measured with STARTRACK (orange line).

3.3. TEPC cluster size distributions: charge collection efficiency in the drift region

In order to study and to verify the expected trend of the charge collection efficiency in the drift region of the TEPC, several distributions at 25 nm were measured with Li-7 ions at different distances between the particle trajectory and the anode wire. Eight measurements between 3 mm (helix position) and 6.25 mm (close to the cathode border) with 0.5 mm steps were performed. From now on, the positions of the irradiations are referred as the relative distance from the centre of the sensitive volume. These irradiations were carried out by moving the TEPC by means of a stepper motor remotely controlled and keeping all the other experimental parameters constant (high voltage of the electrodes, gas pressure, energy and intensity of the beam). For each position, two cluster size distributions were obtained (one measured by the AC-TEPC and one simulated by means of the Monte Carlo code) and the corresponding mean values M_1 were computed.

Some of the acquired ionization cluster size distributions for different distances between the centre of the TEPC drift region and the primary track of the Li-7 ion compared with Monte Carlo simulations are plotted in Figure 7. Once again, the good agreement in terms of both shape and width between measurements and simulations suggests a minor influence of the gas gain variance on the measured cluster size distributions. This is not the case for the point corresponding to 1.5 mm from the centre of the volume which corresponds to a position near the cathode wall. This is ascribable to a wall effect due to the presence of the cathode. In fact, this behaviour is absent in the correspondence of the helix.

Figure 8 shows the comparison between the experimental and simulated mean ionization cluster sizes as a function of the distance *d* between the centre of the TEPC sensitive region and the particle trajectory.

The trend of M_1 is reproduced both by experimental and simulated data: the mean cluster size is maximum in the centre of the sensitive region, while it experiences a steep decrease if *d* approaches the edges of the sensitive region (the helix at 3 mm and the cathode wall at 6.5 mm). It should be stressed that the uncertainties of the experimental data are only statistical.

This final characterization demonstrates that the TEPC response is constant and well reproducible in the interval -0.75 – 0.75 mm. It should be pointed out that the assumption on the hollow cylindrical sensitive region is supported by the behaviour of the cluster size distribution near the cathode and the helix. This is also reproduced by Monte Carlo simulations in which the multiplication region is neglected. The agreement between Monte Carlo simulations and experimental data is very good, except for the point corresponding to d = 1.5 mm, for which the simulation overestimates the M_1 value. This is ascribable to the already discussed wall-effect.

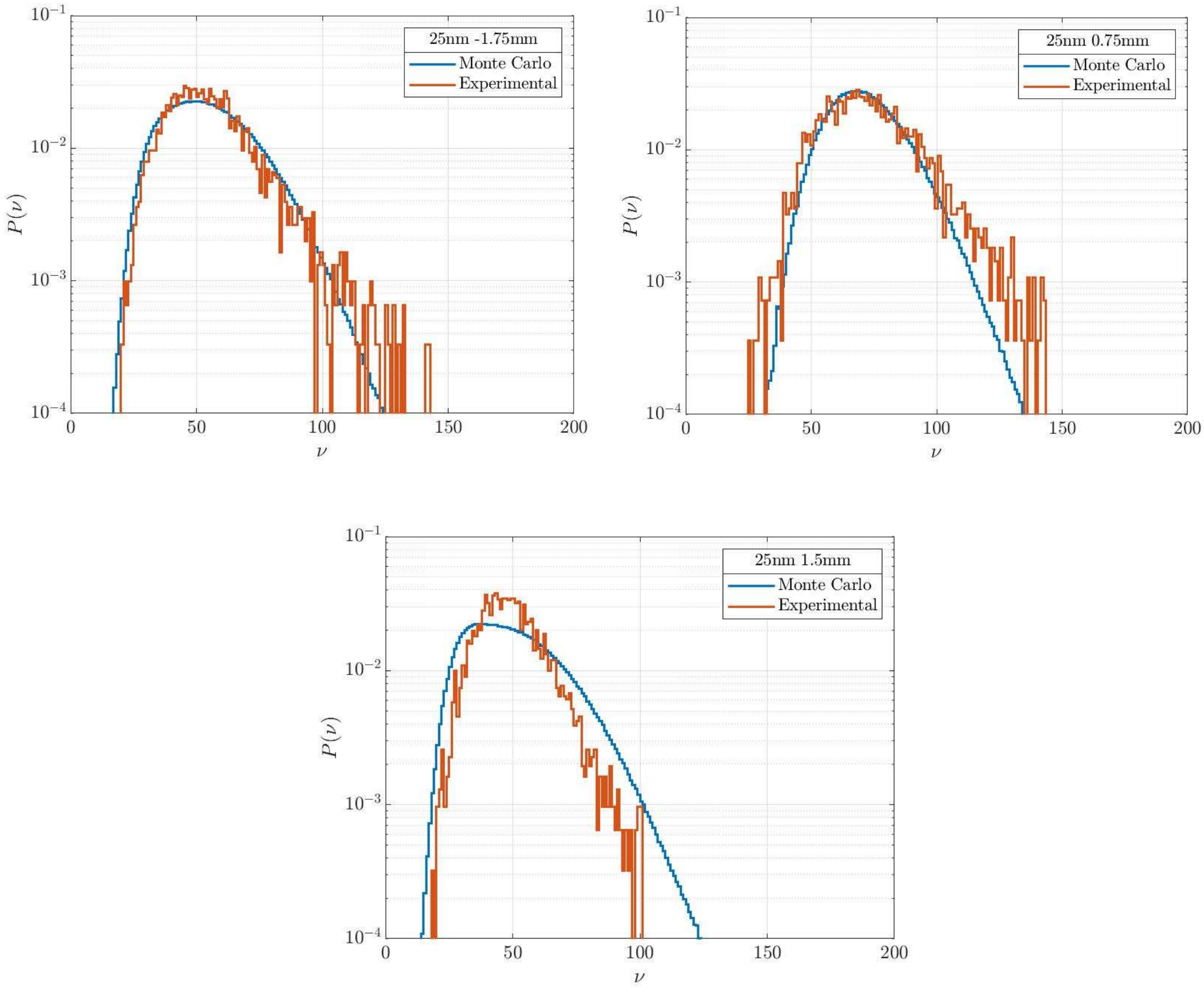

Fig. 7: Experimental (orange dotted line) and simulated (blue continuous line) cluster size probability distributions at 25 nm for different distances between the Li-7 track and the centre of the TEPC drift region.

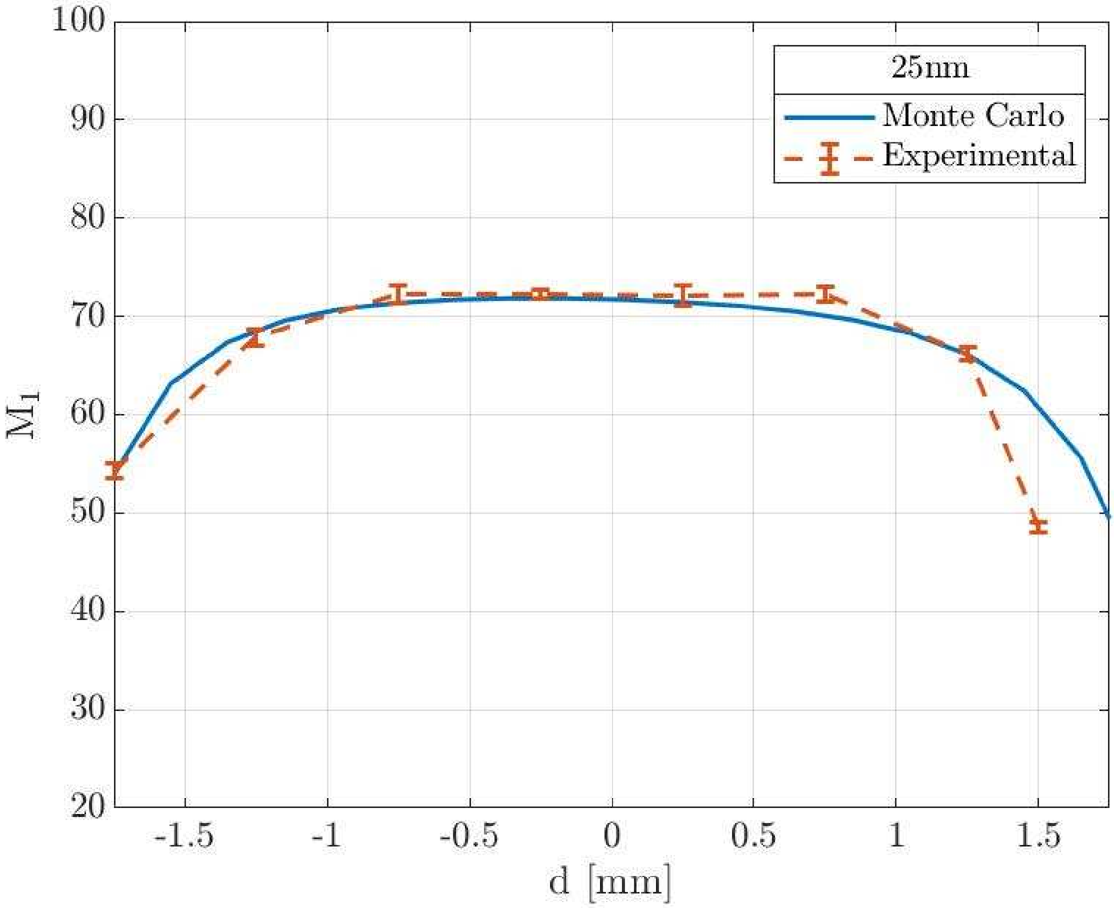

Fig. 8: Experimental (orange dotted line) and simulated (blue continuous line) mean cluster size values at 25 nm against the distance between the Li-7 track and the centre of the TEPC drift region.

Conclusions

A new "open" avalanche-confinement TEPC capable of simulating sites down to 25 has been installed along the STARTRACK nanodosimeter beam for achieving the first direct comparison between ionization cluster size distributions measured with a proportional counter (that includes a variance component due to fluctuations of the avalanche process) and with a single-electron counter.

Experimental campaigns were carried out with He-4 particles from a ^{244}Cm isotopic source and 26.7 MeV Li-7 ions produced by the INFN-LNL Tandem-Alpi accelerator. The calibration of the AC-TEPC with He-4 particles based on Monte Carlo simulations allows to measure the ionization cluster size distributions also for a different radiation quality.

Cluster size probability distributions of 26.7 MeV Li-7 ions turned out to be in good agreement with simulations, which do not include the gas avalanche, and also with cluster size distributions measured with the STATRACK counter, if the detection efficiency of the nanodosimeter is taken into account. This result suggests a minor contribution of the avalanche stochastics to the final shape of the spectra, at least for this radiation quality, and encourages the use of proportional counters for the experimental characterization of therapeutic hadron beams at the nanometer level. Other measurements for sparsely ionizing particles are planned in the next future to confirm this experimental evidence.

Acknowledgements

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