Abstract Probability distributions of the size of ion clusters created in “nanometric” volumes of nitrogen by single $\alpha$-particles of a gold-plated $^{241}$Am source, were measured and compared with those calculated by Monte Carlo methods in the same geometry. The diameter of the sensitive volumes had a mass per area of between 0.015 $\mu$g/cm$^2$ and 1.3 $\mu$g/cm$^2$ which, for a material at unit density, corresponds to a nanometric target volume 0.15–13 nm in diameter. These nanometre sizes were simulated experimentally in a device called the Jet Counter. This consists of a pulse-operated valve which injects into an interaction chamber an expansion jet of molecular nitrogen gas, which is crossed by a narrow beam of $\alpha$-particles. The resulting ions are counted and analyzed from the point of view of ionization cluster formation. The measured or calculated cluster size probabilities prove that the formation of ionization clusters along a “nanometre” track is governed by Poisson’s law only in the case of very small target volumes, due to the contributions by secondary electrons. The present ionization cluster probabilities produced in “nanometric” volumes 0.15–13 nm in diameter, are the first ever determined experimentally and confirmed by Monte Carlo simulation.

Introduction

It is commonly accepted to assume that early damage to genes and cells by ionizing radiation starts with early damage to the DNA molecule. The damage is initiated within the tracks of charged particles in the form of ionization or excitation clusters. Detailed knowledge of cluster formation in targets comparable in size to fragments of the DNA is, therefore, decisive for a deeper understanding of the action of ionizing radiation on the living organism. Because of this fact, experimental and theoretical methods well suited for studying the formation of ionization clusters on a nanometre scale are extremely important in the field of radiation biology and for the development of future measuring devices for radiation protection purposes. After several attempts made since the early 1970s [1, 2, 3, 4, 5] to fulfil this task, the so-called Jet Counter was developed by Pszona and Gajewski [6] which is well suited for measuring ion cluster distributions created by single charged particles in nanometre track sections, in the range of up to 10 nm at unit density. In the Jet Counter, a structure of nanometre size at unit density is simulated by injection of a pulsed gas beam of nitrogen into an interaction chamber. Nitrogen was used because of the knowledge of low-energy electron ranges, which were used for calibration, and of a good set of particle interaction cross sections. For first measurements of the distribution of ion clusters due to $\alpha$-particles in nanometre structures of nitrogen performed with the counter, see the recent publication by Pszona et al. [7]. Parallel to the experiments, the formation of ionization cluster distributions produced by charged particles in the sensitive volume of the Jet Counter was studied by Monte Carlo simulation. The aim of such modeling was firstly, to elucidate the consistency of the calculated and experimental results, secondly to evaluate the overall efficiency of the ion cluster measurements for different nanometre sites and, thirdly to test the deconvolution method applied to the experimental data.

The present paper (i) summarizes the main aspects of the experimental method, (ii) gives a detailed description of the Monte Carlo procedure, and (iii) presents and discusses the most complete results of measured ion cluster distributions due to $\alpha$-particles for a wide range of simulated nanometre sizes at unit density, comparing them with the results obtained by Monte Carlo simulation.
The Jet Counter experiment

As mentioned in the introduction, the Jet Counter developed by Pszona and co-workers was used to measure the probability distribution of the size of ionization clusters caused by α-particles in “nanometric” volumes of nitrogen. The counter consists of a pulse-operated valve which injects into an interaction chamber an expansion jet of molecular nitrogen gas, which is crossed by a narrow beam of α-particles. The resulting ions are counted and analysed from the viewpoint of ionization cluster formation. The mass per area of the jet of nitrogen gas along the particles’ track is between about 0.015 µg/cm² and 1.3 µg/cm² and corresponds to a simulated target size of between 0.15 nm and 13 nm at unit density. The main problems to be solved for performing the measurements were (i) the modeling of the simulated nanometre sizes, (ii) the determination of the efficiency of single ion counting, (iii) the influence of the efficiency of single ion counting on the measurement of ion cluster sizes, and (iv) the measurement of the cluster size distributions. These distributions are defined in the following by the probability distribution of the size of ionization clusters caused by α-particles in “nanometric” volumes of nitrogen.

\[ M(T) = \sum_{v=0}^{\infty} vP(T,v) \]  

As will be shown later for α-particles, the mean ion cluster size \( M(T) \) with respect to nanometric targets is for the most part governed by the particles’ ionization cross section and can, therefore, be used to characterize the radiation quality.

Modeling of simulated nanometre sizes

Similarly to the common microdosimetric technique, where a gas cavity filled with a tissue-equivalent gas is substituted for a mammalian cell, it has been assumed for the present investigation that also sub-cellular target volumes a few nanometre in size at unit density, can be modeled by a gas cavity of appropriate density and size, at least in a first approach. In the Jet Counter, the gas cavity which simulates a nanometric volume at unit density is created by the method explained in part A of Fig. 1: the simulated nanometre size (SNS) is obtained by pulse expansion of nitrogen gas into an interaction chamber (IC), leading to a pulsed jet of nitrogen molecules. The volume of the interaction chamber is of cylindrical shape, 10 mm in diameter and 15 mm in height, with walls made of stainless steel (or tissue-equivalent material if necessary). Pulse expansion is performed by means of a fast piezoelectric valve (PZ), which is connected to a second valve and injects nitrogen through a nozzle with an orifice, 1 mm in diameter, from a reservoir (R) into the IC below the nozzle. The IC is subdivided into two cavities separated by a grid (S), the cavity between S and the open bottom of the IC representing the simulated nanometre-sized site from which ions can be collected with known detection efficiency. This cavity is schematically shown in the enlargement B of Fig. 1 and forms a cylinder with a height equal to its diameter (see part C of Fig. 1).

Two methods could be used, at least in principle, to determine the mass per area of the SNS along its diameter at half the normal height. The first is based on the measurement of the attenuation of a mono-energetic electron beam which diametrically crosses the SNS, and the second is based on the determination of mean cluster sizes produced by α-particles when they penetrate diametrically through the SNS. The positions of the electron gun (EG) and the electron detector (CH1) used in the attenuation experiment are shown in part D of Fig. 1, relative to the positions of the \( ^{241}\text{Am} \) source and the Si detector used for the α-particle measurements.

To give an impression of the results of the attenuation measurements, Fig. 2 shows the time dependence of the nitrogen gas flow through the SNS, represented by the transmission of mono-energetic electrons through the gas layer across the diameter of the cavity, as a function of the time elapsed after operation of the piezoelectric valve PZ. The minimum of the transmission curve represents the maximum nitrogen density within the SNS cavity, which is maintained for about 200 µs. It should be mentioned that the SNS site is only well defined during this time period. To control the instant gas density within the interaction chamber, the gas pressure inside the reservoir R is measured. The mass per area of the gas layer across the diameter of the cavity was determined using the method of Pszona and Gajewski [6], which is based on the expression of Subba Rao [8] for the transmission or absorption of mono-energetic electrons. The parameters controlling the transmission rate are firstly, the pressure of nitrogen in the reservoir R (or the voltage steering the piezoelectric valve PZ) and secondly, the practical range of electrons at a specified energy. For the present purpose, the practical ranges of 100 eV and 500 eV electrons were taken from Grosswendt [9]: a 100 eV electron beam was used for scaling the thickness below 0.2 µg/cm² and a 500 eV electron beam for the range between 0.2 µg/cm² and 1.5 µg/cm².

The second method for the determination of the mass per area of the simulated nanometre structure is based on the fact that the greater part of the mean size of ion clusters is due to the direct ionization of nitrogen molecules by the α-particles whereas the contribution by secondary electrons is of only minor importance. It can be assumed, therefore, that the mean cluster size \( M_\alpha(T) \) due to α-particles at energy \( T_\alpha \) is almost exclusively determined by the ratio \( (\lambda_p)/(\lambda_p)_\text{ion} \) of the mass per area \( \lambda_p \) of the nitrogen layer along the particles’ track and the mass per area \( (\lambda_p)_\text{ion} \) of the mean free path length of α-particles.