Monte-Carlo “step-by-step” simulation of the early stages of liquid water radiolysis: 3D visualization of the initial radiation track structure and its subsequent chemical development

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1. Introduction

The understanding of radiation action on liquid water, an important constituent of cells and living organisms (~85%), is a crucial prerequisite for the accurate description of the effects of radiation on more complex chemical and biological systems. Gel dosimeters, which are used to modelize the biological medium upon irradiation, also have an important water content (~90%) and are another example that requires a detailed knowledge of the complex succession of events that follow absorption in water of ionizing radiation.

Radiation interacts with water molecules mainly by ionization and excitation (“physical” stage). These ionized and excited molecules, and the ejected secondary electrons, will reorganize on the picosecond time scale to generate the primary free radical, ionic and molecular species distributed in small clusters called spurs (“physicochemical” stage). For low linear energy transfer (LET) radiation (such as $^{60}$Co γ-rays or fast electrons), the spurs are separated by large distances relative to their diameter. For high-LET radiation, they overlap to form a continuous cylinder. This initial distribution of radiolytic species, the so-called radiation track structure, is the starting point for the “nonhomogeneous chemical” stage, which extends from $\sim 10^{-12}$ s, that is, the beginning of molecular diffusion, up to $\sim 10^{-6}$ s, the time at which the spur processes are complete. At this time, the species that have escaped from spur reactions become homogeneously distributed throughout the bulk of the solution. The radical and molecular products are then available for reaction with added solutes at moderate concentrations. For an extensive review of the radiolysis of water and aqueous solutions, see ref. [1].

2. Methods and Results

In the calculations reported here, the complete sequence of all individual stochastic events that take place following the irradiation of water is modeled using our Monte-Carlo code IONLYS-SBS [2-4]. This code, which we use to describe the three consecutive, temporal stages (physical, physicochemical, and chemical) of radiation action, follows the reactant trajectories on an event-by-event basis, enables one to keep control of the spatial distribution and identity of all generated radical and molecular species ($e_{aq}^\cdot$, $\cdot$OH, $H^\cdot$, $H_2$, $H_2O_2$, $H^+$, $OH^-$, $HO_2^-/O_2^-$,...) at all times. At present, our “step-by-step” program is able to simulate ion and electron track structures and their subsequent chemical development for various LET varying from $\sim 0.3$ to 1000 keV/μm. It also takes account of the possibility of multiple ionization of water at high LET. As an example, Fig. 1 shows the initial,
nonhomogeneous deposition of radiation energy in water exposed to 24-MeV helium ions (LET ~ 26 keV/µm). As we can see, the track consists of a cylindrical “core” produced by the ion track itself and a surrounding “penumbra” of ejected, comparatively low-LET, secondary electrons (“β-rays”).

The three-dimensional expansion of the track structure shown in Fig. 1 is displayed in Fig. 2. This figure shows the time-dependent cross sections of the nonhomogeneous spatial distributions of each of the radiolytic species (e\textsuperscript{-aq}, OH\textsuperscript{-}, H\textsuperscript{+}, H\textsubscript{2}, and H\textsubscript{2}O\textsubscript{2}) in liquid water exposed to 24-MeV helium ions from 1 ps to 10 µs. The track geometry is the same as that in Fig. 1, but here the track segment is viewed end-on over the XZ plane (see ref. [3]).
the radiolytic species produced, calculated with our code IONLYS-SBS from 1 ps to 10 μs. As can be seen, these radial distribution profiles of species essentially reflect the trajectories of the ejected δ-rays. At an early stage of track development, the $\epsilon_{aq}$ and $\cdot$OH distributions are far denser than those of the other radiation-induced reactants (H, H$_2$, and H$_2$O$_2$). This observation is consistent with the much greater radiolytic yields (or $G$-values, representing the numbers of species formed or destroyed per 100 eV of deposited energy) found for these two radical species; in fact, for 24-MeV $^4$He$^{2+}$ ions, our calculated values of $G(\epsilon_{aq})$ and $G(\cdot$OH) at 1 ps are, respectively, ~4.2 and 5.0 molecules/100 eV, while the corresponding yields of H, H$_2$, and H$_2$O$_2$ are only ~0.69, 0.33, and 0.18 molecules/100 eV, respectively [3]. Figure 2 also clearly reveals that the initial ion track structure is gradually lost as time progresses because of the diffusion of the species, and has completely disappeared by about 1 μs.

In order to offer a better understanding of radiation action on more complex chemical and biological aqueous systems, our code enables a user, in a 3D environment and in real time, to either visualize the spatial distribution of all reactive species present in the track at a chosen simulation time, or to present an animation of the chemical development of the radiation track over a chosen time interval.

3. Conclusions
As polymer gel dosimeters consist basically of a hydrogel in which monomers are dissolved and in view of their potential use as 3D dosimeters in radiotherapy, we believe that our simulations could play a useful role in the development of our knowledge of the underlying physical and chemical processes that take place in those systems upon irradiation, and in the optimization of their chemical composition in terms of both dose and spatial accuracy.

4. References
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