



Simulation of ion-induced water radiolysis in different conditions of oxygenation



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ABSTRACT

We have investigated the production of free radicals induced by swift ions during the radiolysis of oxygenated water and analyzed the underlying mechanisms in detail. To this aim, we simulated, by Monte-Carlo, the irradiation of water by projectiles with LET values ranging from 1 to 300 keV/μm for a partial pressure of oxygen in air from 0 to 750 mmHg, and for times up to 10 μs after ion impact. For low-LET radiation, we observed an increase in production of (HO₂ + O₂⁻) with oxygen pressure and a saturation. At 1 μs, the saturation occurred at a pressure of 20–30 mmHg and the maximal yield amounted to 0.3 μmol L⁻¹ per Gray. For the same conditions, we observed similar trends for high-LET ions, but we observed a significant reduction in the yield values and an attenuation of the saturation behavior.

By underlining similarities between the yield of (HO₂ + O₂⁻) and the oxygen effect observed in radiobiology, we discuss the role of (HO₂ + O₂⁻) in oxygen effect and suggest a general mechanism for this phenomenon.

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1. Introduction

Water radiolysis concerns many fields, including safety in nuclear reactors, nuclear waste storage, radioprotection, space exploration, radiotherapy and more recently hadrontherapy with light ions. It has therefore been studied for many years both experimentally and theoretically. The production of radicals depends not only on the irradiation dose but also on the radiation characteristic (particle, energy). Such dependence is due to the spatial distribution of the radicals produced, for which the heterogeneity increases with the Linear Energy Transfer (LET). As a consequence, no standard model based on homogeneous chemistry can be applied. Instead, models of heterogeneous chemistry need to be considered and generally consist in a detailed description of diffusion and chemical reaction processes for the set of chemical species produced by irradiation. It is also well-known that radical production by water radiolysis can be greatly modified by addition of a solute to pure water. Among others, the solvation of oxygen molecules in water can significantly modify the radiolysis process.

In fact, we know that a major effect is the production of O₂⁻/HO₂ radicals through the following reactions:



The effect of this solute is the most interesting of all when oxygen is a major constituent of air. Studying the effect of dissolved oxygen on water radiolysis at various partial pressures of oxygen in air may therefore lead to a better understanding of water radiolysis in aerated conditions.

To our knowledge, few experiments [1,2] have been performed to quantify the production of O₂⁻/HO₂ radicals at various concentrations of dissolved oxygen and for various LET values. This is surprising since such an evaluation could help to understand, for instance, the effect of oxygen in radiobiology. Indeed, it has been shown that cell sensitivity to ionizing radiation is affected by the partial pressure of oxygen in air and among other mechanisms, the production of superoxide anions has been suggested as contributing to cell death [3–6,1]. Although biological systems are extraordinarily complex and differ significantly from the radiolysis of oxygenated water, a better knowledge of the role of dissolved

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oxygen in water radiolysis could provide a firmer ground to understand the oxygen effect in radiotherapy.

From a theoretical point of view, one can cite the work by Tomita et al. [7] and Watanabe and Saito [8] for which the study was limited to low-LET radiation. To our knowledge, no systematic calculation of O_2^-/HO_2 as a function of the concentration of oxygen and for various LET values has yet been published. This study therefore aims at simulating the production of radicals during the radiolysis of oxygenated water by swift ions. We consider projectiles with a LET value ranging from 1 to 300 keV/ μm , a partial pressure of oxygen in air from 0 to 750 mmHg and a time of observation up to 10 μs after ion impact.

We first give a brief description of the simulation. Then, we present the results showing the effect of dissolved oxygen on the time-dependent yields for the main species. After that, we focus on the yield of O_2^-/HO_2 radicals and present their evolution with the partial pressure of oxygen for irradiations by proton, carbon and argon ions at various LET values. These results are first analyzed and discussed from a chemical point of view and the discussion is extended to the domain of radiobiology.

2. Materials and methods

Our simulation aims at predicting the number and spatial distribution of free radicals and molecules produced in water by ionizing radiation. The radiation can be a beam of high-energy photons, electrons or ions. In the present work, it consisted in simulating the interaction of an ion characterized by its charge (proton, carbon and argon) and its energy (10–65 MeV/n). The water sample was infinite in the perpendicular direction to the ion trajectory (see Fig 1). The sample thickness was sufficiently small to assume that the ion energy was constant but thick enough to reduce edge effects (no species are indeed produced beyond each extremity of the track segment, except for δ -electrons). In fact, the thickness ranged from 5 μm for the highest LET ion (Ar [65 MeV/n]) to 1500 μm for the lowest LET (H [65 MeV/n]). In these conditions, the number of simulated chemical species amounted to approximately 2.5×10^5 species at the first step of the simulation. In addition, we applied periodic boundary conditions, so that any molecule produced outside the sample or that crossed one of the surfaces, was immediately re-injected in the sample by translation (see Fig. 1).

The principle of our simulation model is largely based on previous work [9–12]. A detailed description can be found in [13,14].

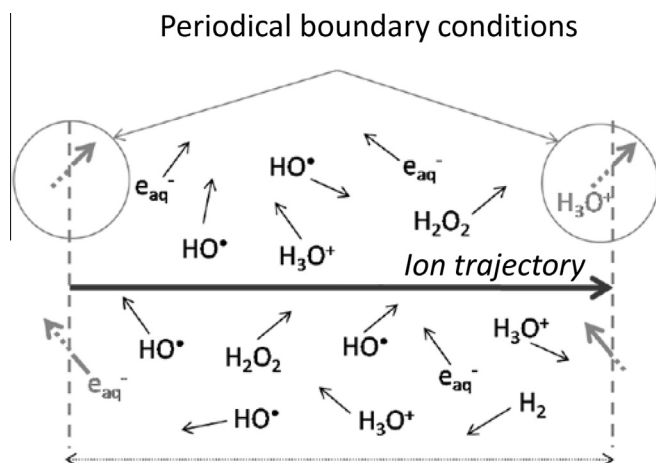
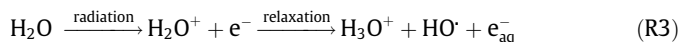


Fig. 1. Sketch illustrating the simulation geometry and boundary conditions. The arrows illustrate the diffusion process during one time step. Any species that cross the boundary (dashed arrow) are re-injected in the simulation volume on the opposite side.

The simulation was divided in three stages: physical, physico-chemical and chemical. First, the *physical stage* was described by Monte Carlo simulations (LQD). The passage of the incident ion generated a spatial distribution of excited (H_2O^*) or ionic (H_2O^+) water molecules. Multiply-ionized species H_2O^{q+} could also be created by swift ions. The ejected electrons (known as primary electrons) were simulated in the same way but the processes of elastic diffusion, electron attachment, and phonon creation were also included. The electrons were simulated until their energy fell to a cut-off energy set at 33 meV (energy of thermalization for a temperature of 300 K). The geminate recombination process was also simulated. The simulation output was a spatial distribution of low-energy electrons (thermalized electrons), ionized and excited water molecules. Considering the concentration of dissolved oxygen molecules, the fraction of oxygen molecules with regard to water molecules is low (less than 1 over 40,000) and ionization of oxygen molecules was neglected.

Secondly, the *physicochemical stage* describes the relaxation of the medium. The thermalized electrons were solvated. The ionized water molecules could either undergo a dissociation process, producing new molecules, or capture an electron to go to an excited state. The excited water molecules could also dissociate, and when dissociation occurred, the fragments were separated by a specific distance. If multiple-ionization took place, atomic oxygen was produced. The dissociation-channel details and the associated probabilities are available in [14]. In the simulation, these relaxation channels were sampled by a random process. The outcome of the simulation was a spatial distribution of chemical species. The major species are hydroxyl radicals, hydronium ions and solvated electrons, and they mainly issued from the single ionization of water molecules according to the channel:



Finally, the *chemical stage* describes the evolution of the spatial distribution of the chemical species with respect to time. Standard chemical models could not be applied since the spatial distribution can be very heterogeneous (see Fig 2). We therefore developed a Monte Carlo simulation [14], very similar to Independent Reaction Time (IRT) [10]. This simulation decomposes the whole evolution into a series of short time steps [10,12]. During each step, the reaction probability was calculated for each pair of species as a function of diffusion constant, charge, reaction rate, reaction spin and radius of the species [10,12]. A random process selected a subset of these reactions by sampling their reaction probabilities. Since the time step was small, most of the species did not react and simply diffused according to a standard diffusion model. The list of reactions and the associated parameters are given in Table 1 and are mainly obtained from [12]. The outcome of the simulation was the time-dependent radical yield defined as the number of chemical species produced per 100 eV of energy transferred to the sample.

We extended the simulation to quantify the effect of different concentrations of dissolved oxygen molecules on water radiolysis. However, as directly introducing oxygen molecules to the simulation would dramatically increase the computing time, we applied the approximation introduced by Green et al. [15,16]. This approximation consists in representing the distribution of oxygen molecules as an homogeneous and inexhaustible reservoir. Such an approximation has already been considered in a simulation of the oxygen effect at low values of LET [17,8] as well as for the study of pH effects [18]. The probability that a specie reacts with the oxygen reservoir during the period of time t can be written as:

$$P(t) = 1 - \exp(-\alpha(x^2 + y^2)) \quad (1)$$

where:

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