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Geant4-DNA simulation of the pre-chemical stage of water radiolysis and its impact on initial radiochemical yields

Wook-Geun Shin^{1,2,3}, Jose Ramos-Mendez⁴, Ngoc Hoang Tran¹, Shogo Okada⁵,
Yann Perrot⁶, Carmen Villagrasa⁶, and Sebastien Incerti¹

¹UMR 5797, Bordeaux University, CNRS, CENBG, 33170 Gradignan, France

²Department of Radiation Oncology, Seoul National University Hospital, 03080 Seoul, Korea

³Biomedical Research Institute, Seoul National University Hospital, 03080 Seoul, Korea

⁴Department of Radiation Oncology, University of California San Francisco, San Francisco, 94143 CA, USA

⁵KEK, 1-1, Oho, Tsukuba, Ibaraki 305-0801, Japan

⁶IRSN, Institut de Radioprotection et de Sûreté Nucléaire, BP17, 92262 Fontenay-aux-Roses, France

Abstract

This paper demonstrates the impact of the pre-chemical stage, especially the dissociation scheme and the associated probabilities, on water radiolysis simulation using the Geant4-DNA Monte Carlo track structure simulation toolkit. The models and parameters provided by TRACs have been collected and implemented into Geant4-DNA. In order to evaluate their influence on water radiolysis simulation, the radiochemical yields (G-values) are evaluated as a function of time and LET using the “chem6” Geant4-DNA example, and they are compared with published experimental and calculated data. The new pre-chemical models lead to a better agreement with literature data than the default pre-chemical models of Geant4-DNA, especially for $\cdot\text{OH}$ radicals and H_2O_2 . The revised chemistry constructor “G4EmDNAChecker_option3” is available in Geant4-DNA version 10.7.

1. Introduction

The mechanistic simulation of biological effects induced by ionizing radiation at the sub-cellular and DNA scales is a scientific challenge. Such an approach could lead to a better understanding of carcinogenesis at low doses and to the development of more accurate risk models [1]. Numerous Monte Carlo track structure (MCTS) codes have been developed so far to predict DNA damage induction. Among them, Geant4-DNA [2-5], PARTRAC [6], TRACs (TRACELE and TRACPRO) [7], and others (see comprehensive reviews in [8-10]), can predict both direct and

indirect damage, from physical interactions and water radiolysis products, respectively. At the sub-cellular scale, it is reported that the contribution of indirect damages becomes more important than that of direct damage below a linear energy transfer (LET) of 300 keV/ μm . [11]. Therefore, a careful modeling of water radiolysis in the cellular medium (liquid water in these codes) is essential.

The "pre-chemical" stage of water radiolysis links the "physical" and "chemical" stages and determines the initial radiochemical yields, called G-values, which are the number of molecular species normalized per 100 eV of deposited energy. During the pre-chemical stage, the excited and ionized water molecules, H_2O^* and H_2O^+ , are dissociated into molecular species according to the pre-chemical model. To date, it is unfortunately almost impossible to experimentally probe the pre-chemical stage on the femtosecond scale. Therefore, it is still common in radiochemical studies to adjust the pre-chemical parameters to match the simulated radiochemical yields to the experimental data, due to the lack of a complete mechanistic understanding of the pre-chemical processes. For example, PARTRAC uses the pre-chemical model of TRACs with modifications of the dissociation scheme and probabilities to reproduce the experimental yields of chemical species. On the other hand, the pre-chemical model of Geant4-DNA (version 10.7) is derived from PARTRAC without any adjustment, but additional physico-chemical processes such as electron attachment and electron-hole recombination are added. These additional processes, indeed more realistic, induce a significant disagreement which makes it difficult to match the experimental data for radiolysis of water.

Thus, in this study, we propose to revise the pre-chemical dissociation scheme and probabilities of the 10.7 version of Geant4-DNA to improve the agreement with the experimental and simulated G-values found in the literature, as a function of time and LET.

2. Materials and methods

2.1 Pre-chemical models

Table 1. Dissociation scheme and associated probability for each channel used in this work, and available in Geant4-DNA, PARTRAC and TRACs MCTS codes. Values in italic indicate changes compared to Geant4-DNA 10.7 [12].

		Channel	Probability (%)			
			This work	Geant4-DNA [12]	PARTRAC [6]	TRACs [7]
Ionization	H_2O^+	$\text{H}_3\text{O}^+ + \cdot\text{OH}$	100	100	100	100
	Auger effect H_2O^{2+}	$2\text{H}_3\text{O}^+ + \text{H}_2\text{O}_2$	<i>100</i>	-	-	100
Excitation	A^1B_1	$\text{H}^+ + \cdot\text{OH}$	65	65	65	65
		H_2O	35	35	35	35
	B^1A_1	$\text{H}_3\text{O}^+ + \cdot\text{OH} + \text{e}_{\text{aq}}^-$	<i>50</i>	55	55	50
		$\text{H}^+ + \cdot\text{OH}$	<i>25.35</i>	-	-	25.35
		$\text{H}_2 + \text{O}(^1\text{D})^{\text{a}}$	<i>3.25</i>	15	15	3.25
		$\text{H}_2 + 2\cdot\text{OH}^{\text{c}}$	<i>$\text{H}_2 + 2\cdot\text{OH}^{\text{c}}$</i>	$\text{H}_2 + 2\cdot\text{OH}^{\text{c}}$	$\text{H}_2 + 2\cdot\text{OH}^{\text{c}}$	$\text{H}_2 + \text{H}_2\text{O}_2^{\text{d}}$
		$2\text{H}^+ + \text{O}(^3\text{P})^{\text{b}}$	<i>3.9</i>	-	-	3.9
		H_2O	<i>17.5</i>	30	30	17.5
	Rydberg, Diffusion bands	$\text{H}_3\text{O}^+ + \cdot\text{OH} + \text{e}_{\text{aq}}^-$	50	50	50	-
		$\text{H}^+ + \cdot\text{OH}$	-	-	-	-
		H_2O	50	50	50	-
Electron capture	Electron attachment H_2O^-	$\text{OH}^- + \cdot\text{OH} + \text{H}_2$	100	100	-	100
	Electron-hole recombination H_2O^*	$\text{H}^+ + \cdot\text{OH}$	<i>35.75</i>	55	-	35.75
		$\text{H}_2 + \text{O}(^1\text{D})$	<i>13.65</i>	15	-	13.65
		$\text{H}_2 + 2\cdot\text{OH}^{\text{c}}$	<i>$\text{H}_2 + 2\cdot\text{OH}^{\text{c}}$</i>	$\text{H}_2 + 2\cdot\text{OH}^{\text{c}}$	-	$\text{H}_2 + \text{H}_2\text{O}_2^{\text{d}}$
		$2\text{H}^+ + \text{O}(^3\text{P})$	<i>15.6</i>	-	-	15.6
		H_2O	<i>35^{\text{e}}</i>	30	-	35^{\text{e}}

^a Oxygen atom in the singlet D state.

^b Oxygen atom in the triplet P state.

^c $\text{O}(^1\text{D}) + \text{H}_2\text{O} \rightarrow 2\cdot\text{OH}$ from Burns et al., 1981 [13].

^d $\text{O}(^1\text{D}) + \text{H}_2\text{O} \rightarrow \text{H}_2\text{O}_2$ from Taube, 1956 [14].

^e Same relaxing probability for B^1A_1 excitation except auto-ionization.

The dissociation scheme of ionized water molecules (H_2O^+) is clearly established [15], $\cdot\text{OH}$ and H_3O^+ , regardless of the ionization level. Furthermore, in this work, the dissociation of H_2O^{2+}

induced by the Auger process is newly implemented in Geant4-DNA. The excited water molecule (H_2O^*) is dissociated according to the five excited states. The A^1B_1 state represents the excitation of the 5th electronic layer ($1b_1 \rightarrow 4a_1$), while the B^1A_1 state corresponds to the excitation of the 4th layer ($3a_1 \rightarrow 4a_1$), and the other states represent Rydberg series and diffusion bands.

As shown in Table 1, Geant4-DNA uses the pre-chemical model of PARTRAC to describe the dissociation scheme after excitation, and considers two additional processes (after electron capture) that are not considered by PARTRAC: electron attachment and electron-hole recombination. Thus, in this study, we applied the B^1A_1 dissociation channel proposed by TRACs [7]. Note that the different atomic oxygen states (^1D or ^3P) are considered as different species in the water radiolysis simulation, and we decided to use the more recent dissociation channel of $\text{O}(^1\text{D})$ into two hydroxyl radicals proposed by Burns et al. [13] instead of that of Taube [14] (which considers dissociation into H_2O_2) in order to obtain better agreement with experimental data.

The dissociation of H_2O^* after electron-hole recombination is accounted for in both TRACs and Geant4-DNA. For this process, an empirical dissociation channel was proposed by Rowe et al. [16], in which it is reported that the fraction of the $\text{H}^+ + \cdot\text{OH}$ dissociation channel is 55% of the species produced. However, the 10.7 version of Geant4-DNA directly uses this value without reducing it by considering the relaxation probability. Therefore, we corrected the dissociation probabilities of electron-hole recombination taking into account those of relaxation probability, for example 35.75% for $\text{H}^+ + \cdot\text{OH}$ channel ($55\% \times 65\%$) [7], for consistency with the original paper by Rowe et al. [16].

In summary, this study includes Auger-generated H_2O^{2+} dissociation, dissociation channel modifications for B^1A_1 excitation, and the electron-hole recombination process.

2.2 Simulation configuration

The public version of Geant4-DNA (version 10.7) and the recently implemented independent reaction times (IRT) technique (available in the example “chem6”) [17, 18] are used in this study to simulate G-values for incident electrons, protons and alpha particles. The simulation configurations and physics settings validated in our previous water radiolysis study [19-22] are used, namely the G4EmDNAPhysics_option2 physics constructor with the ELSEPA electron elastic scattering model and the Meesungnoen electron thermalization model. For radiolysis simulation, we choose the G4EmDNAChemistry_option3 chemistry constructor, which uses the same chemistry parameters as RITRACKS from Elliot's work [23, 24], and the proposed modification of the pre-chemical parameters presented in this work.

Computational and experimental data sets presented by [25-39] are used for comparison with the simulated G-values as a function of time. For G-values as a function of LET, computational and experimental data obtained by [22, 40-45] are compared with the results. It should be noted that most of the experimental data are measured under arbitrary control of the solvent concentration, whereas the simulations are performed for pure liquid water.

3. Results and discussion

3.1 G-values as a function of time

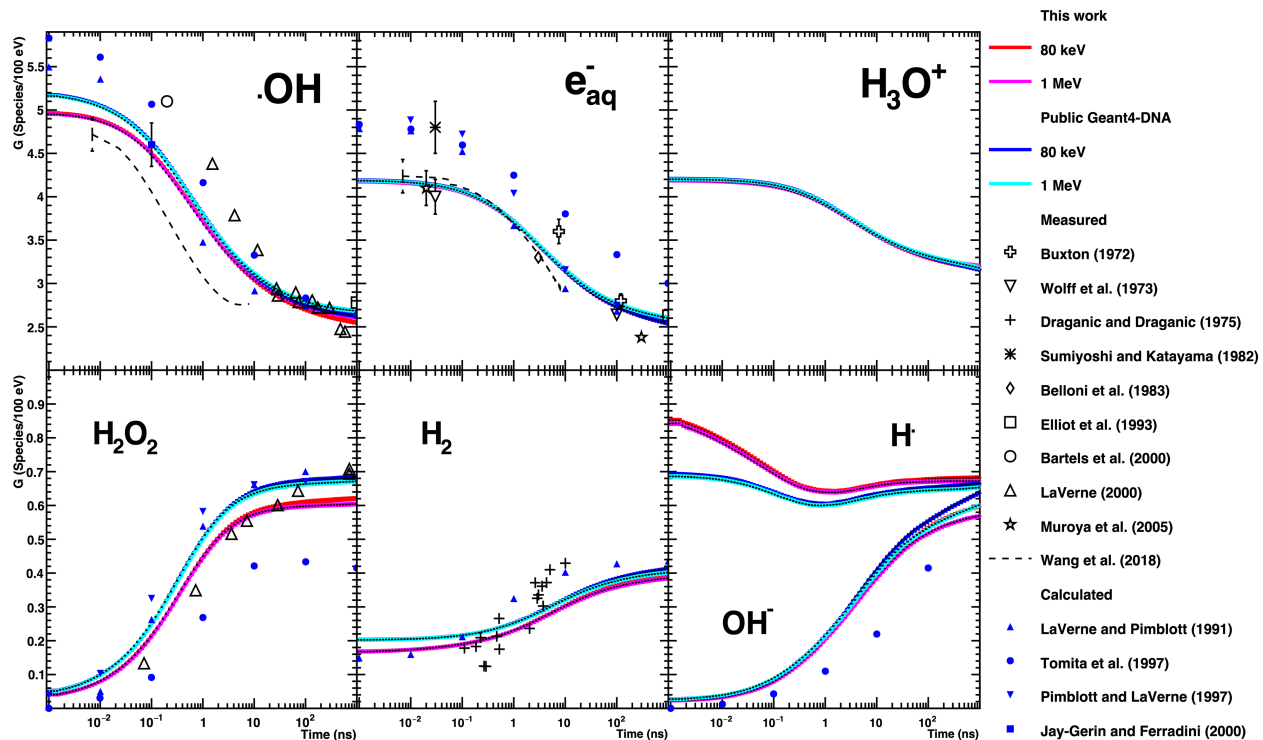


Figure 1. The time evolution of G-values for incident electrons calculated with the modifications proposed in this work (red and magenta curves) and by the 10.7 version of Geant4-DNA (blue and cyan curves), compared to the measured and calculated yields. For Wang et al. (2018), the fit data are shown.

We first present in Figure 1 the simulated radiochemical yields as a function of time, using Geant4-DNA 10.7 (blue and cyan curves) and the improved pre-chemical stage proposed in this work (red and magenta curves). The LET values of all calculated and measured references are between 0.186 - 0.314 keV/ μm . Thus, only the results for 80 keV and 1 MeV electrons, whose LET values are 0.61 and 0.16 keV/ μm , respectively, are shown in Figure 1.

The G-values are not much affected by the LET changes in the low LET region, for example, between the 0.16 and 0.61 keV/ μm curves. Thus, we can conclude that the variation observed in the experimental data comes mainly from the different experimental setups. The red and magenta curves have lower G-values for $\cdot\text{OH}$ radicals, H_2O_2 , H_2 , and OH^- than the blue and cyan curves. The G-values of these species calculated in this study are closer to recent experimental data such

as those of Wang et al. [39] (dashed line, for $\cdot\text{OH}$ and e_{aq}^-), measured at the shortest time after irradiation (7 ps), within the experimental uncertainty.

For H_2 molecules, the initial G-value with the new pre-chemical parameters is 0.16 molecules/100 eV at 1 ps. This value is closer to the experimental estimate of Schwarz et al. [46] reported at 0.15 molecules/100 eV (shown as the initial value of LaVerne and Pimblott [30]).

We observed that the differences between both values on $\cdot\text{OH}$ and H^\bullet radicals saturate at a late time. However, up to 5 ns (which is the proposed time limit of the damage simulation in Geant4-DNA [47]), the G-values of $\cdot\text{OH}$ radical (which are mainly responsible for indirect DNA damage) decrease and the two models differ by up to 5%.

3.2 G-values as a function of LET

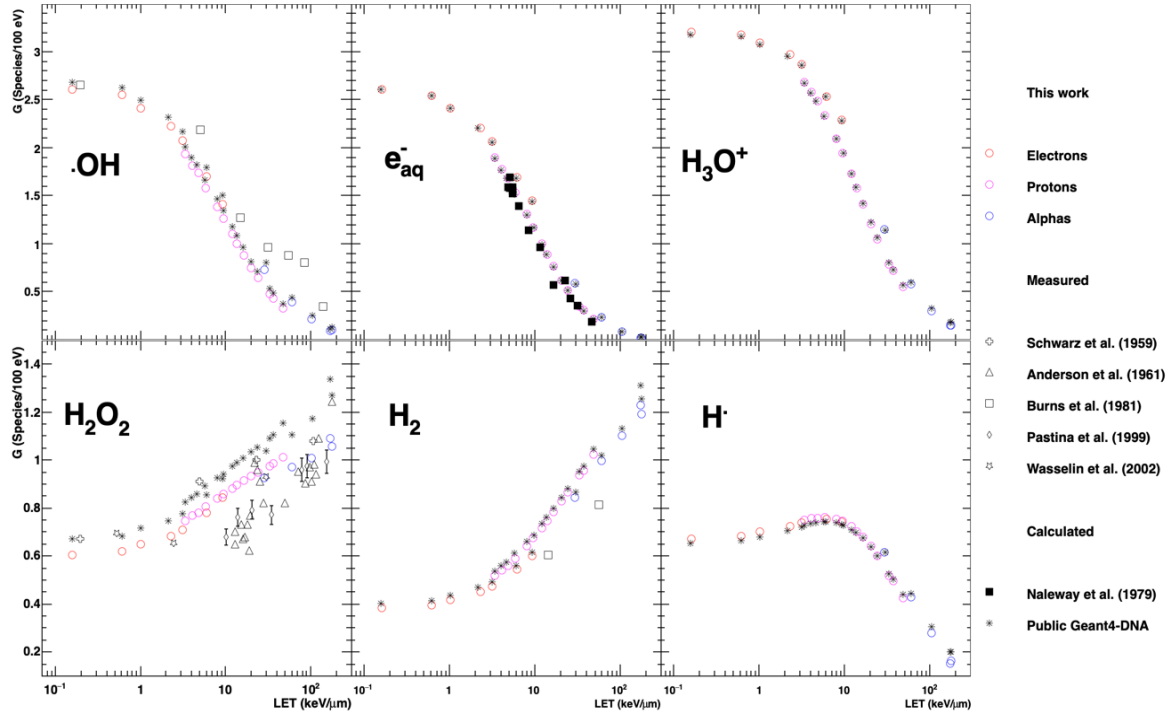


Figure 2. The G-values at the end of the chemical stage (1 μ s) as a function of LET for incident electrons, protons and alphas (open coloured circles), compared with measured and calculated yields.

Next, G-values as a function of LET in the range of 0.16 - 177.85 keV/ μ m are calculated at 1 μ s, as shown in Figure 2. We can observe a slight change for \cdot OH, H₂, and H \cdot due to the correction of dissociation probabilities in the recombination process and B¹A₁ excitation. The main improvement here is that the G-values of H₂O₂, which previously showed higher values than literature data using the 10.7 version of Geant4-DNA, are smaller due to the decrease in the initial number of \cdot OH radicals at the beginning of the chemical stage.

4. Conclusion

In this study, the pre-chemical stage of Geant4-DNA is evaluated by comparing it with the

TRACs MCTS codes. The Geant4-DNA dissociation scheme is then revised to improve agreement with experimental and computational G-values as a function of time and LET found in the literature.

The new dissociation scheme including B^1A_1 excitation and the electron-hole recombination process directly influences the G-values of $\cdot\text{OH}$, H_2 , and H^\cdot species. Our results are close to recent experimental data, especially for $\cdot\text{OH}$ and H_2O_2 generated by $\cdot\text{OH}+\cdot\text{OH}$ reactions. This adjustment could reduce the Geant4-DNA simulated indirect DNA damage yields, which were relatively higher than experimental data [18]; this will be the subject of another study. The new pre-chemical model presented in this work will be soon released in Geant4-DNA as an alternative option.

In future work, the dissociation of H_2O^+ cation [48] will be improved and multi-ionized water molecules [49] will be added based on recent literature for the accurate modeling of pre-chemical stage.

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