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ARTICLE

Modeling Radiation Chemistry in the Geant4 Toolkit

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Simulation of biological effects of ionizing radiation at the DNA scale requires not only the modeling of direct damages induced on DNA by the incident radiation and by secondary particles but also the modeling of indirect effects of radiolytic products resulting from liquid water radiolysis. They can provoke single, double strand breaks and base damage by reacting with DNA. The Geant4 Monte Carlo toolkit is currently being extended for the simulation of biological damages of ionizing radiation at the DNA scale in the framework of the "Geant4-DNA" project. Physics models for the modeling of direct effects are already available in Geant4. In the present paper, an approach for the modeling of radiation chemistry in pure liquid water within Geant4 is presented. In particular, this modeling includes Brownian motion and chemical reactions between molecules following water radiolysis. First results on time-dependent radiochemical yields from 1 picosecond up to 1 microsecond after irradiation are compared to published data and discussed.

KEYWORDS: Geant4, Geant4-DNA, DNA, microdosimetry, liquid water, chemistry, radiolysis, radiobiology, radiation damage

I. Introduction

The accurate modeling of interactions of ionizing radiation with biological matter remains a challenge in radiobiology. Monte Carlo simulation techniques can provide solutions allowing for example the prediction of biological DNA damages, such as single, double strand break and base damages, which can be measured experimentally. Such damages may directly impact on the cell mutation, quiescence or death. The Geant4 Monte Carlo simulation toolkit, an open source set of object-oriented libraries developed for modeling the passage of particles through matter, covers a large variety of application domains, ranging from high energy physics to space and medical applications. In particular, the Geant4-DNA project,[1,2] an activity of the Geant4 collaboration, aims at providing Geant4 with new open source capabilities specific to radiobiology applications, including the modeling of elementary physical interactions down to the sub-electronVolt scale, liquid water radiolysis and radiation chemistry. The project focuses primarily on the prediction of early direct and non-direct DNA damages up to 1 μs after irradiation.

Advanced simulation codes have already been developed by several groups for the modeling of ionizing radiation damages on the DNA molecule. They include radiation che-

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mistry modeling (e.g., the PARTRAC software,[3,4,5] the work of S. Uehara and H. Nikjoo[6] and the RADACK software[7]) and some of them also include a detailed DNA geometrical model in order to study DNA strand damages and structure modification. A detailed review of these codes is available in[8]. However, these codes are usually not easily accessible to users and they are often designed for very specific applications which can limit their applicability domain. As an alternative, the Geant4-DNA project is based on the general purpose Geant4 Monte Carlo toolkit which offers to users a rich set of electromagnetic, optical and hadronic physics processes from the electronVolt range up to a few GeV/n, which can be combined in a multi-scale approach.[9] In parallel Geant4 proposes sophisticated geometry modeling capabilities for example recently applied from the scale of biological cells to the scale of planets.[10-11] The platform is available freely on the Internet for several computing environments.[12]

Potential applications of the Geant4-DNA project are foreseen for long duration manned space exploration programs as well as for the use of radiation in the medical domain (e.g., particle therapy) where radiation effects at the cellular level are of concern.

This work presents the first version of a prototype software developed in the framework of the Geant4-DNA project for the modeling of liquid water radiation chemistry in the Geant4 toolkit. The aim of this prototype is to demonstrate the possibility to deliver to the scientific community such modeling capabilities through a general purpose and open source Monte Carlo simulation toolkit.

II. Principle of Radiation Chemistry Simulation

One of the potential applications of radiation chemistry simulations for cellular radiobiology is to evaluate the ratio between DNA strand breaks created by primary and secondary ionizing particles directly on the DNA molecule (the so-called "direct effects") and those provoked by reactive oxygen species (the so-called "indirect effects") produced through water radiolysis by incident ionizing radiations on the biological medium, mainly liquid water.

After ionization or excitation by a primary or secondary particle, a water molecule can dissociate into new molecules. These new molecular species can either interact amongst themselves, producing other molecules, or react with the DNA molecule and finally induce DNA damages. Consequently, after the physical irradiation, the number of molecules of a given species in liquid water changes with time.

In order to verify the reliability of such a simulation, the time-evolution of the number of molecules in liquid water (without simulating explicitly the DNA molecule itself) can be compared to other theoretical predictions. For a given molecular species, this evolution depends on the molecules' capacity to diffuse into the medium (pure liquid water in our case) and depends also on the proximity of the molecules with their respective potential reactants.

III. Scenario of Radiation Chemistry in Liquid Water

The stochastic Monte Carlo simulation of early biological damages on the DNA molecule from ionizing radiation follows several identified consecutive stages, as explained globally in References 4, 7 and 13.

1. The Physical Stage

The first stage following cellular irradiation is the so-called *physical stage*. In this stage, all physical interactions take place. For e.g. in the case of electrons, all the elastic and inelastic interactions are simulated with Geant4-DNA, namely, the elastic scattering, the ionization and the electronic excitation.

2. The Physico-Chemical Stage

From 1 femtosecond (fs) to 1 picosecond (ps), the *physico-chemical stage* encompasses the very fast events (mainly electronic), such as thermalization and solvation of subexcitation electrons, electronic hole migration and fast electronic recombination that leads to chemical bond breaks. For the prototype development reported in this paper, we follow the approach proposed by PARTRAC,[4,5] where branching ratios and thermalization distances are the same for any incoming particle and energy.

Excited water molecules are assumed to decay at 100 fs after irradiation.[13]

The electronic holes of ionized water molecules quickly migrate from a molecule to another. This process lasts about 0.5 fs.[14,15]

At 10 fs after the electron removal,[16] a proton transfer occurs between the final ionized water molecule and a nearby water molecule to originate H_3O^+ and •OH molecules from the reaction: $H_2O^+ + H_2O \rightarrow H_3O^+ + \bullet OH$.

The secondary electrons produced during the *physical stage* get thermalized within a characteristic time of 110 fs through vibrational excitation of water and they become solvated within a characteristic time of 250 fs.[17] In our simulation, the subexcitation electrons can undergo dissociative attachment, vibrational excitation and elastic scattering processes until they reach 25 meV. Afterwards, we consider them as solvated and they start to diffuse through Brownian motion.

The hot dissociation fragments of water molecules are assumed to become all thermalized within 1 ps. At 1 ps, the placement of the fragments from the hit mother water molecule can drastically affect the simulated radiochemical yields. If, for example in the case of a water molecule in the A1B1 excitation state, the dissociation fragments H• and •OH are placed too close from each other, they would rapidly recombine into H_2O.

3. The Chemical Stage

From 1 ps to 1 microsecond (μs), in the so-called *chemical stage*, the molecular species can diffuse through the medium and interact with each other or with the DNA mo-

lecule. The diffusion process and mutual interactions are described in more details in Section V.

IV. Software Requirements

In order to simulate radiation chemistry of liquid water in Geant4, the software fulfills the following five requirements (SR):

- SR#1: description and management of molecules, including some of their static (name, number of atoms, ground state configuration, decay table…) and dynamic (electronic configuration, diffusion coefficient…) properties;
- SR#2: identification of the electronic state of excited and ionized water molecules in the corresponding Geant4-DNA physics models;
- SR#3: decay process for molecules following excitation and ionization, including thermalization;
- SR#4: diffusion process for molecules according to Brownian motion;
- SR#5: chemical interaction process between diffusing molecules;

Geant4 9.4 does not currently include any class for handling molecules. With this prototype, it is the first time that Geant4 is extended to track molecules and model mutual interactions between them. Note that the prototype software described in this work is compatible with Geant4 9.4 release and will be delivered publicly with default input parameters for the modeling of water radiolysis.

V. Molecular Species and Chemical Processes

Under ionizing radiation, excited and ionized water molecules may decay and dissociate into new molecules (e^-_{aq} [a], H_2, H•, •OH, H_3O^+) which can diffuse and interact mutually to produce other molecules (OH^-, H_2O_2). These e^-_{aq}, H•, •OH and H_2O_2 species can also directly interact with DNA components.

New prototype processes in Geant4 for liquid water radiolysis and the resulting chemistry have been developed in Geant4 in the framework of the Geant4-DNA project. We have adopted the approach followed by the state-of-the-art Monte Carlo PARTRAC software, which is described in detail by their authors in the literature.[4,5] Consequently, in this work, we pay a particular attention to the time-dependent radiochemical yields calculated with PARTRAC. The adopted software design was made flexible enough to allow in the future the inclusion of alternative radiochemistry models.

With the present software prototype, the *chemical stage* can be simulated up to 1 µs after irradiation. The simulation starts with the positions of radiolytic products of the water molecules at 1 ps after the physical irradiation, using the branching ratios adjusted by PARTRAC.[4]

[a] e^-_{aq}: a solvated electron in water (also named aqueous solvated electron) is a free electron in an *aqueous* solution surrounded by a "shield" of water molecules.

The hot products have reached a so-called "thermalization distance" from the hit mother water molecule before starting a Brownian motion. The method adopted to simulate the thermalization process in this work can be found in.[4] The flexibility of the prototype software allows the user to easily implement his/her own thermalization distance computation method for each decay channel.

Brownian diffusion of radiolytic products uses a random change of direction after a time step Δt, corresponding to a geometrical mean step size given by:

$$\langle R \rangle = \sqrt{6.D.\Delta t}$$

where D is the diffusion coefficient. The diffusion coefficient depends on the molecular species and on its excited state, while the time step can be selected as a function of the physical time.

The criterion for the chemical reactions to occur is the physical distance between two molecules. If two molecules are closer than a calculated reaction radius,[18] related to the chemical reaction rate, a reaction is assumed to happen, using a jump-through correction.[18]

VI. Simulation Parameters

The default branching ratios (**Table 1**), diffusion coefficients (**Table 2**), reaction rates (**Table 3**) and time steps (Δt) (**Table 4**) of this prototype software are those proposed by PARTRAC,[4] but the user still has the possibility to easily define his/her own parameters and models.

Table 1 Branching ratios of a water molecule at 1 ps as described in Reference 4

Electronic state	Decay Channel	Fraction (%)
All ionization states	H_3O^+ + •OH	100
Excitation state A1B1: (1b1) → (4a1/3s)	•OH + H•	65
	H_2O + ΔE	35
Excitation state B1A1: (3a1) → (4a1/3s)	H_3O^+ + •OH + e^-_{aq}	55
	•OH + •OH + H_2	15
	H_2O + ΔE	30
Excitation state: Rydberg, diffusion bands	H_3O^+ + •OH + e^-_{aq}	50
	H_2O + ΔE	50

Table 2 Diffusion coefficients for the diffusing species as described in Reference 4

Species	Diffusion coefficient D (10^{-9} m^2 s^{-1})
e^-_{aq}	4.9
•OH	2.8
H•	7.0
H_3O^+	9.0
H_2	4.8
OH^-	5.0
H_2O_2	2.3

Table 3 Reaction rates as described in Reference 4

Reaction	Reaction rate (10^{10} M^{-1} s^{-1})
$H\bullet + e^-_{aq} + H_2O \rightarrow OH^- + H_2$	2.65
$H\bullet + \bullet OH \rightarrow H_2O$	1.44
$H\bullet + H\bullet \rightarrow H_2$	1.20
$H_2 + \bullet OH \rightarrow H\bullet + H_2O$	4.17×10^{-3}
$H_2O_2 + e^-_{aq} \rightarrow OH^- + \bullet OH$	1.41
$H_3O^+ + e^-_{aq} \rightarrow H\bullet + H_2O$	2.11
$H_3O^+ + OH^- \rightarrow 2\ H_2O$	14.3
$\bullet OH + e^-_{aq} \rightarrow OH^-$	2.95
$\bullet OH + \bullet OH \rightarrow H_2O_2$	0.44
$e^-_{aq} + e^-_{aq} + 2\ H_2O \rightarrow 2\ OH^- + H_2$	0.50

Table 4 Time steps Δt with respect to the physical time, as described in Reference 4

Time interval (s)	Δt (ps)
Until 10^{-11}	0.1
$10^{-11} - 10^{-10}$	1
$10^{-10} - 10^{-9}$	3
$10^{-9} - 10^{-8}$	10
Above 10^{-8}	100

VII. Time-Dependent Radiochemical Yields

For a given molecular species, the time-dependent radiochemical yield G is defined as the number of molecules produced for a total absorbed energy of 100 eV in the irradiated medium:

$$G = \frac{N(t)\times 100}{E\ (eV)},$$

where $N(t)$ is the number of molecules and E is the total energy deposit by the incident ionizing particle into the medium, expressed in eV.

Experimental data on time-dependent chemical yields are rare and the simulations often do not reproduce the exact conditions of the experimental set-up (type and energy of the incoming particle, volume of the target). However, in order to fill in this lack, most authors compare their simulated results with experiments at similar LET value.

In order to verify our prototype software, we compare our results with other simulation codes (PARTRAC[4,5] and Uehara's and Nikjoo's work[6]). However, as Geant4-DNA and PARTRAC[4,5,19] physics models are not identical, the outcome of the physical stage in the two simulations will necessarily differ. The physics models adopted in Geant4-DNA are fully described in Reference 2.

The presented results were obtained using the above default parameters, by shooting 2.5×10^4 incident electrons of 1 MeV and considering only the first 10 keV lost by the primary track, as done in PARTRAC[4] and Uehara's and Nikjoo's work.[6] All secondary particles were tracked.

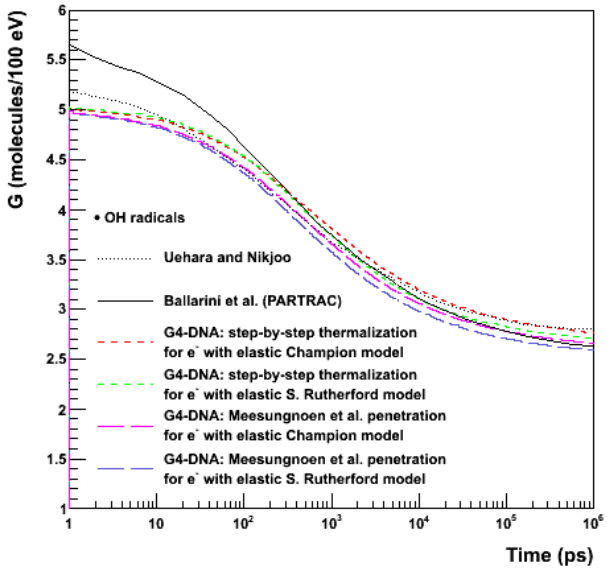

Fig. 1 Prototype results on •OH radiochemical yields (molecules/100 eV) from 1 MeV incident electrons with respect to time (in picosecond). References: Ballarini *et al.* (PARTRAC),[3] S. Uehara and H. Nikjoo.[6]

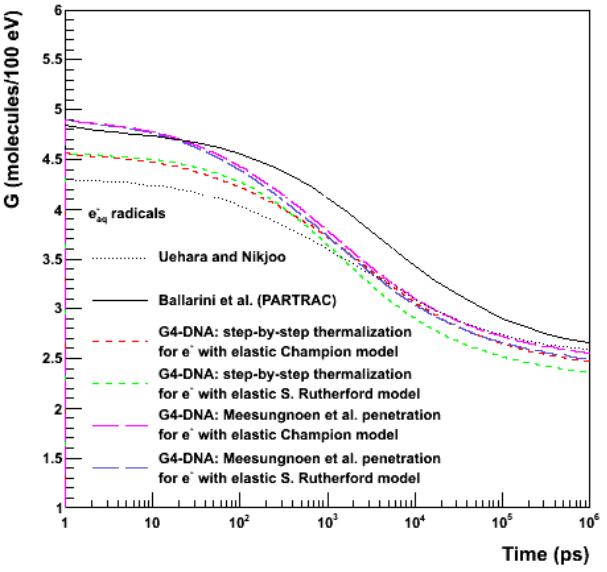

Fig. 2 Prototype results on e^-_{aq} radiochemical yields (molecules/100 eV) from 1 MeV incident electrons with respect to time (in picosecond). References: Ballarini *et al.* (PARTRAC),[3] S. Uehara and H. Nikjoo.[6]

The resulting radiochemical yields for •OH and solvated electrons are shown in **Figs. 1** and **2** respectively. These preliminary results have been obtained using the following Geant4-DNA processes (the corresponding Geant4-DNA models are indicated in parenthesis): electron elastic scatter-

ing (Champion model or Screened Rutherford model), electronic excitation (Born excitation model), ionization (Born ionization model), vibrational excitation (Sanche model) and dissociation attachment (Melton model). These processes and models are available in the Geant4 release 9.4 and they are described in References 2 and 20.

The results of this prototype software are compared to other theoretical calculations. The computation of the radiochemical yields was performed using two different computation methods for the tracking of very low energy electrons. In the first approach, we followed the electrons step-by-step down to 25 meV using the processes mentioned above. In the second approach, we used the penetration for low energy electrons computed by Meesungnoen *et al*.:[21] when the electron reaches a kinetic energy inferior to the lowest excitation level (8.22 eV), it is displaced in one unique and final step whose mean value corresponds to a fit of the penetration curve given in[21] and with an isotropic direction. In the two approaches described above, electron elastic scattering was computed using either the Champion model or the Screened Rutherford model.[2]

Compared to PARTRAC, Geant4-DNA early yield for hydroxyl radicals appears slightly underestimated. The use of different physics models between PARTRAC and Geant4-DNA could account for this difference. One can also note that the dissociation scheme of B1A1 excitation channel is slightly different in Ballarini *et al*.[3] with respect to that of Kreipl *et al*.[4]

On the other hand, radiochemical yields at 1 microsecond for both solvated electrons and hydroxyl radicals appear to converge in all simulations. However, one can notice that the step-by-step tracking of very low energy electrons leads to smaller G-values than the approach which uses the one step placement of the electron. The slight shift observed at 1 ps for solvated electrons can be explained considering that below the excitation threshold, the electron interacts with the liquid water medium through three processes: elastic scattering, dissociative attachment and vibrational excitation. In the step-by-step approach, the electron can undergo dissociative attachment down to 4 eV, which depresses the number of solvated electron present at one picosecond. However, in the second approach, no absorption of the electron via dissociative attachment occurs below the lowest excitation energy.

Furthermore, the thermalization distance computed at 7 eV via the step-by-step approach with the Champion (Screened Rutherford) elastic model is about 40% (60%) lower than the Meesungnoen's approach. As a result, in the first approach, the secondary electrons are closer to the water molecule they come from and thus closer to the products of dissociation. As a consequence, in the case of full tracking of low energy electrons, albeit there are less solvated electrons at 1 picosecond (because they are captured by water molecules), reactions are more likely to occur at earlier times due to their smaller computed range during the physics stage.

Although the comparison of PARTRAC physics models with Geant4-DNA ones is out of the scope of this contribution, we have verified as an illustration that a systematic 100% increase of the electron cross sections for ionization and excitation leads to no significant increase of G values at 1 ps and to a 10% increase on both yields at 1 μs in the described conditions.

VIII. Conclusion

A first software prototype dedicated to the simulation of liquid water radiation chemistry has been developed within the general purpose and open source Geant4 Monte Carlo simulation toolkit, in the framework of the Geant4-DNA project. This prototype demonstrates the feasibility of such modeling in Geant4 and will allow to users to benefit fully from Geant4 capabilities and features. The comparison to other liquid water radiation chemistry simulations, although highlighting some differences, shows a qualitatively good agreement. The described prototype software is still under development and uses recent models presented in Reference 20 which are included in the Geant4 9.4 release. It will be fully released publicly in the near future and will be validated against experimental data. We also expect to compare our results to dedicated radiolysis experiments that will be performed at the Laboratoire de Radiolyse (CEA Saclay, Gif-sur-Yvette, France) in order to measure time-dependent •OH yields by using chemiluminescence technics.[22, 23]

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