Comparison among different Geant4-DNA physics models of proton transportation in nano-layers

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***Abstract*—In this paper are presenting a comparison by using experimental data about chemical damage for all possible physics list for Geant4-DNA (Geant4 version 10.02.p01) for protons incident beam interacting with homogeneous nano-layers of water. Associated to the chemical damage it were evaluated: radial profile of energy deposition, profile of energy deposition on depth, number of interactions, proportion among possible processes and stopping power of the incident particle. The application considers water ultra-thin layers of thicknesses from 2 nm to 200 nm with monodirectional and monochromatic (2 MeV to 20 MeV) protons impinging normally on its entrance surface. It was evoked the standard physics list ("G4EmDNAPhysics" class) and other 5 physics list available (names Opt 1 up to 5). Concerning the experimental data, the films were bombarded by 2 MeV H+ in vacuum at a HVEE 3 MV Tandetron and X-ray photoelectron spectroscopy was performed on the irradiated samples at Université de Namur, Belgium. The preliminary results show that radial and depth profile of energy deposition to all physics lists evaluated presented as expected, with exception for the physics list Opt1. This physics list shows a peak of energy deposition at the end of the layer on depth profile. On radial profile it is clear that the secondaries electrons transported by Opt1 presented larger range than the presented by other evaluated physics list. It was observed for few simulations considering standard physics list that for thickness above ~350 nm the curve of depth profile achieve it "saturation" and stop the increasing on energy deposition with depth.**

***Index Terms*—Geant4-DNA, Monte Carlo methods, proton transportation, ultra-thin layer.**

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[1] Introduction

The Geant4 [1-3] is a general-purpose particle-matter Monte Carlo simulation toolkit. In the version Geant4-DNA, it was extended for modeling the early biological damage induced by ionizing radiation at cellular and sub-cellular scale [4-6]. This simulation scale enables modeling the interaction of particles with matter at nanoscale. However, validation of Geant4-DNA results against experimental data is still limited and it is important to take into account that one needs to be careful on generalizing the simulation results. In this paper, results of simulations of MeV protons impinging on homogeneous nano-layers of water, using different physics lists (including beta versions for Geant4-DNA running on version 10.02.p01) are being compared. It includes the radial and depth profiles of the deposited energy, the number of interactions, the proportion among possible simulated processes and the stopping power of the incident particle. Besides that, an indirect validation will be presented using experimental data on chemical damage cross-sections of 2 MeV H on PMMA and PVC thin films.

[2] Methodology

The strategy used to evaluate the reliability/accuracy of data was defined to evaluate the expected behavior for specific known quantities and compare the experimental chemical damage to the chemical damage calculated based on standard thermally activated model[7] and the simulated radial energy deposition. The known quantities used to compare were: energy deposition as function of the distance from the center of core (named as radial profile), energy deposition as function of the depth (named as depth profile) and the stopping power as function of ultra-thin layers for different energy of incident proton beam.

A. The Monte Carlo simulation

The application developed in Geant4-DNA version 10.02.p01 is based on a simple geometry, considering an ultra-thin layer of water in a semi-infinite configuration with protons impinging normally to the entrance surface. Different thicknesses of water ultra-thin layers were simulated (2 nm, 4 nm, 6 nm, 8 nm, 10 nm, 20 nm, 30 nm, 40 nm, 50 nm, 100 nm and 200 nm). The proton beam was simulated monodirectional and monochromatic, with

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initial kinetic energies of 2 MeV, 5 MeV, 10 MeV, and 20 MeV. The class "G4EmDNAPhysics" (henceforth called Std) is the stable and recommended physics list to be evoked by Geant4-DNA. It was defined as the reference for the comparison made with the other physics lists available. For each ultra-thin layer and beam energy, 10^5 histories were run, considering 1 nm of cut-off for secondary particle generation. According to the Geant4-DNA official webpage the "Geant4-DNA processes are all discrete; as such, they simulate explicitly all interactions and do not use any production cut, so this 1 nm cut will have no effect on the Geant4-DNA Physics results"[1]. The Std and the other 5 available physics lists (named Opt 1 up to 5) were evoked. During the transportation, in the sensitive volume, for each interaction the follow information was recorded: pre-step and post-step position (x, y and z in nm); kinetic energy of the particle (in MeV); deposited energy due to the interaction (in MeV); event identification; identification of the parent particle; track identification; step identification; process identification and particle of interaction. At the end of the run it was calculated and printed out traceability information: physics list evoked, total number of histories, number of histories per hour, total absorbed energy and its statistical fluctuation, real running time, and the system and user running times.

Later this information was organized/accumulated on bins representing depth and radial profile and counting the number of hits. The total energy profile deposited in each ultra-thin layer was used the estimate the stopping power for protons.

B. The experimental setup and chemical damage estimation

In the experiments with the polymer thin films, high-grade poly(methyl methacrylate) (PMMA) and poly(vinyl chloride) (PVC) powder were dissolved and spun onto polished Si wafers. Homogeneous thin films, with thicknesses from 4 to 200 nm, and very low roughness (~0.3 nm RMS) were obtained. The films were bombarded by 2 MeV H+ in vacuum at a HVEE 3 MV Tandetron (Porto Alegre, Brazil) with a set of fluences ranging from 10^{14} up to 2.8×10^{15} ions/cm^2. X-ray photoelectron spectroscopy (XPS) was performed on the irradiated samples at Université de Namur, Belgium, to evaluate bond-breaking cross-sections of C-O and C-Cl bonds as a function of the thickness of the polymer.

[3] RESULTS

Figures 1 to 4 show examples of the results observed in the simulation. Figure 1 shows example of radial and depth profiles of deposited energy to standard physics list, both profiles present a behavior similar to the expected, with the radial profile presenting the $\frac{1}{r^n}$, where n ~2, but slightly larger than 2, as was reported by [8] on his validation for Geant4 to radial profiles for Geant4.

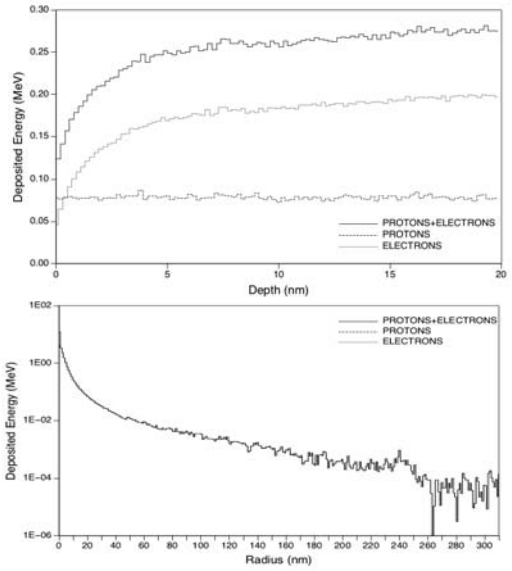

Figure 1: Example of depth (left plot) and radial profile (right plot) considering protons of 2 MeV passing through 20 nm water layer by evoking standard (stable) physics list.

Figure 2 presents the visual comparison among different models evoked for transportation. This is an example of the behavior observed to all energies and ultra-thin layer thickness. All models presented similar behavior to the Std (stable) models, with exception for the physics list Opt 1. For this model, the depth profile showed a peak at the end of the layer (on exit surface of the primary particles of the water layer). In the radial profile, it is clear that the particles (especially the secondary electrons) have deposited less energy on Opt1 than the other evoked physics list, presenting a larger range for the secondary electrons. Further investigation and statistical analyses are needed to generalize these results. The expected behavior for radial profile was observed being observed for $\frac{1}{r^n}$ behavior, where n is in a range of 2.1 and 2.28.

The results for different initial kinetic energies showed profiles with similar behavior, however with a lower amount of deposited energy. Considering we are evaluating layers of water with thicknesses up to 200 nm, and the range of the secondary electrons is larger than that (for the investigated energies), the total deposited energy increases slightly with layer thickness. It was observed in few simulations using the standard physics list that, for thicknesses above ~350 nm, the curve of the depth profile achieves its expected saturation value close to the tabulated stopping power.

1 http://geant4-dna.in2p3.fr/styled-3/styled-9/index.html

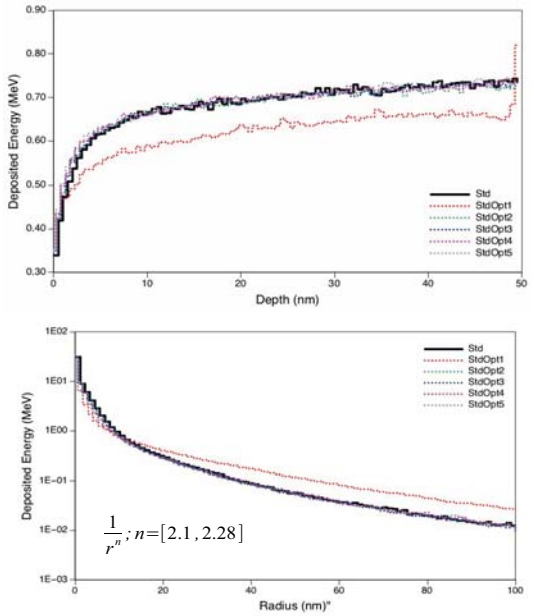

Figure 2: Example of comparisons among different physics lists evoked on depth (left plot) and radial (right plot) profiles considering protons of 2 MeV passing through 20 nm water layer.

The Fig 3 presents examples of the behavior of the number of interactions for protons and electrons to all evaluated physics list. One need to take in mind that the increase on number of interactions represents an increase on running time.

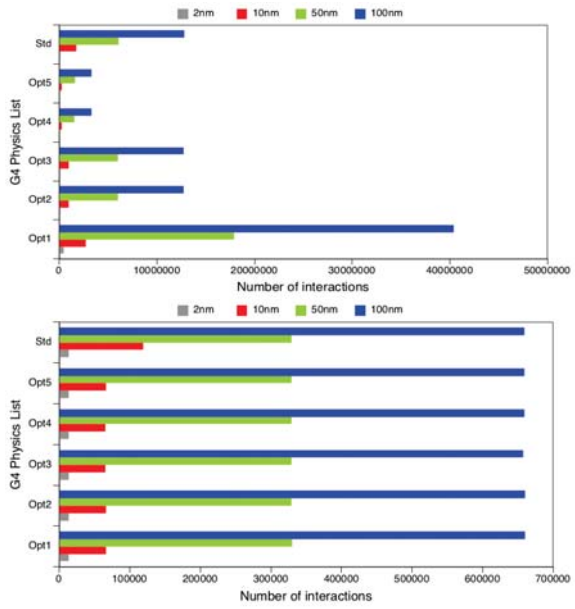

Fig 3: Number of interactions for electrons (top) and for protons (bottom) considering incident protons of 10 MeV and different thicknesses of ultra-thin layers.

It is easy to observe that for the energy range we are simulating the protons present no significant change on number of interactions. In the other hand, the electrons presented significant changes. Taking the Std physics list as reference one can see that Opt1 presented the larger number of interactions for electrons. Considering the whole dataset simulated, all thicknesses and energies evaluated, the Opt1 presented 1.5 to 4.1 time more interactions than Std. The Opt2 and Opt3 presented similar behavior having 0.5 to 1.0 time the number of interactions of Std. The Opt4and Opt5 presented 0.15 to 0.35 time interaction than Std. The similar behavior presented by Opt2 and Opt3 and by Opt4 and Opt5 were expected due the evoked process for electrons in the energy range evaluated.

The Fig 4 presented the stopping power behavior as function of the ultra-thin layer to all evaluated energies.

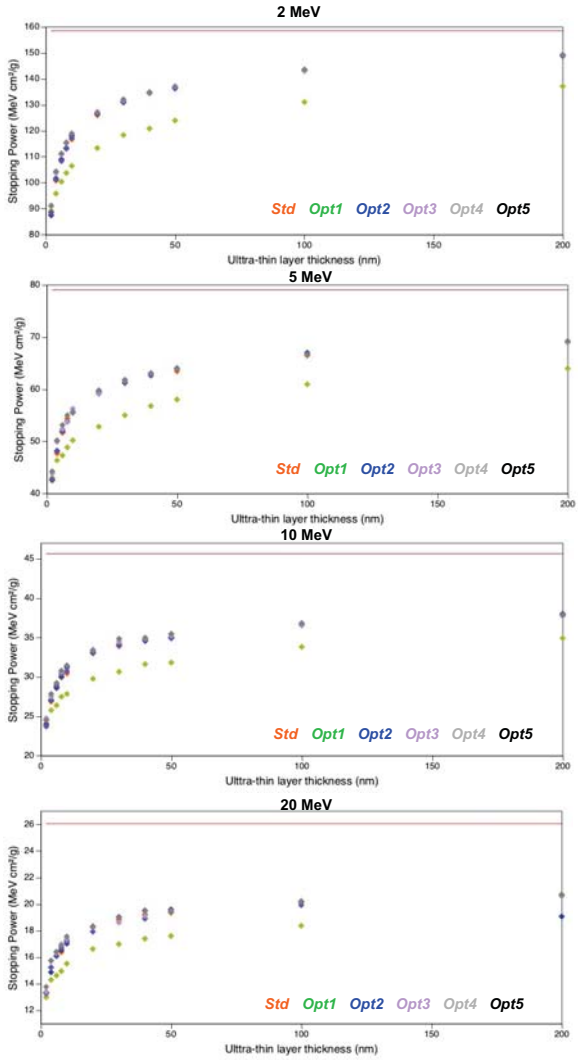

Fig 4: Stopping power behavior as function of the ultra-thin layer for different incident energy: 2 MeV, 5 MeV, 10 MeV and 20 MeV. The red line represents the expected value of stopping power published

All the values of stopping power based on simulated deposited energy are sub-estimated, as expected since the thickness of the ultra-thin layers are inferior to the range of electrons this behavior was expected. According our observations of tendencies the stopping power for 2 MeV incident protons will achieve the (macroscopic) value published on stopping power and range table for proton – PSTAR[2] - of National Standards and Technology. To

2 https://physics.nist.gov/PhysRefData/Star/Text/programs.html

other energies evaluated 5 MeV, 10 MeV and 20 MeV the following differences were observed respectively 8%, 10% and 18%. One can see that the percentage difference increase with the increase on energy of the incoming beam. It was expected since the probability of interaction reduced with the increase on incident beam energy. We are aware of the limitations of this comparison, since the values published by PSTAR are macroscopic measurements. Since we didn't find stopping power data collected in the same conditions we are simulating, our strategy was to compare the data considering PSTAR value and a limit to the tendency curve. Values above it would be considered inconsistent for the simulation taking into account that ultra-thin layers have it thickness smaller than the range for electrons, which means that non of all energy deposited is stopped in the volume of interest.

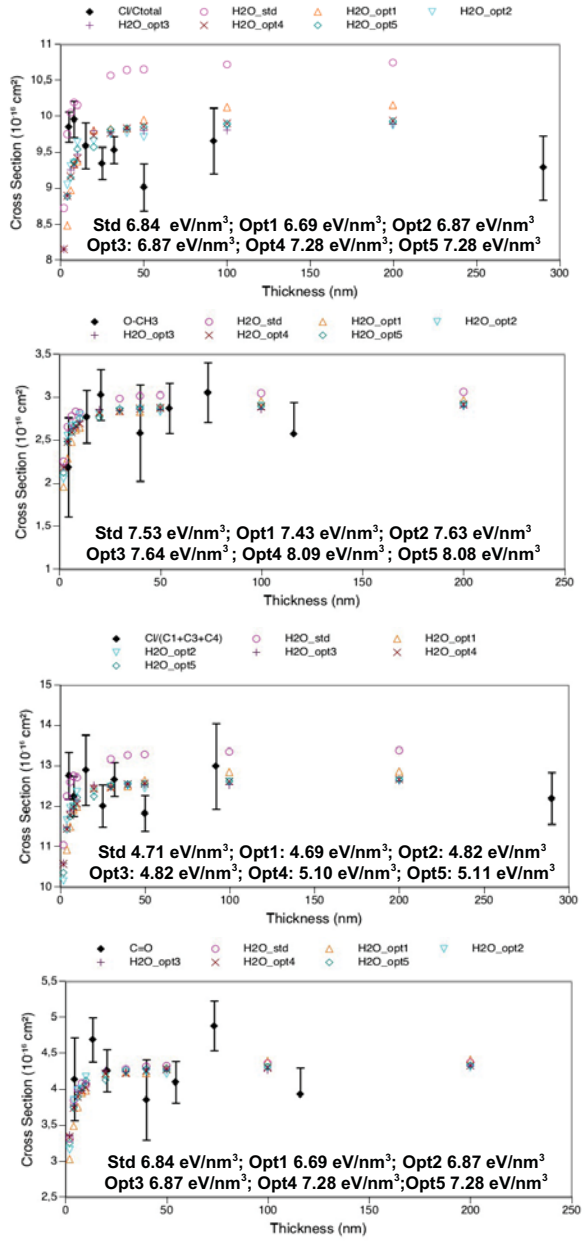

Fig 5: Chemical damage for different physics list with activation energy density to evaluate different bond-breaking.

The Fig 5 presented the chemical damage (experimental and simulated data) as well the activation energy density to different bond-breaking. According to [9] and adapting to our conditions, the activation energy to get a relabel results must be in the range of 1 eV/nm^3 and 10 eV/nm^3.

In a general analyses, considering the chemical damage, all models presented good agreement to this methodology defined. The Opt1 presented always the low value for the activation energy density.

[4] Conclusions and Suggestions

It was observed for different evaluated physics lists and simulated energies, that the models and processes evoked by physics list Opt1, presented 1.5 up to 4.1 times more interactions than the one using the Std physics list, increasing also the running time. In general, no significant differences were observed for the total deposited energy among all models, with exception for Opt1 which presented inconsistencies in the profile curves with a non-expected behavior.

Evaluating the energy deposition profiles in depth and radial and taking standard physics list as reference, one can observe that: (i) the Opt2 to Opt5 presented similar results with percentage differences on simulated values lower than 6%; (ii) the Opt1 presented in depth profile one peak at the end of the energy deposition profile and on radial profile presented one significant change on curve shape (worst results for fitting to $\frac{1}{r^n}$).

In a general analyses the radial deposited energy decreases systematically for thinner layers. Considering the running time, differences were observed associated to the number of interactions (specifically electrons) among the different physics lists evoked. On this evaluation the Opt1 presented longer running time and larger number of interaction and Opt4-5 presented the lower running time and lower number of interactions.

The simulated stopping power always presented lower values than the NIST values. In this way it was Opt1 the physics list that generate the lowest stopping power values. In general the stopping power values increase with the increase of thickness of the ultra-thin layers.

The evaluation of chemical damage cross section (based on radial energy deposition profile) have shown that the bin size influences on the behavior of the damage cross section curve. These results have shown good agreement between the experimental chemical damage cross-sections in the polymer films and the extracted values calculated from the simulated radial energy density profiles in water, despite the differences in the materials used.

We are planning to activate the chemical processes and simulate the chemical damage in conditions similar to experimental data in a future paper.

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