Step-by-step simulation of radiation of radiation chemistry using Green Functions for diffusion-influenced reactions

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Introduction

- The irradiation of biological systems leads to the formation of radiolytic species such as H, OH, H₂, H₂O₂, eaq, etc. [1]
- These species react with neighboring molecules, which result in damage in biological molecules such as DNA.
- Radiation chemistry is therefore very important to understand the radiobiological consequences of radiation [2].
- In this work, we discuss an approach based on the exact Green Functions for diffusion-influenced reactions which may be used to simulate radiation chemistry and eventually extended to study more complex systems, including DNA.

Green functions

- The exact Green functions for an isolated pair are known analytically [3-4]:

\[
\begin{align*}
\text{Green Functions:} & \\
& \text{Free particles: } E_0(x,y) \text{ and } W_0(x,y) \\
& \text{Bound particles: } E_1(x,y) \text{ and } W_1(x,y) \\
& \text{Product: } E_2(x,y) \text{ and } W_2(x,y)
\end{align*}
\]

- The coefficients α, β and γ are related to the reaction rate constants (k0=4πRD):

\[
\begin{align*}
\alpha &= \frac{1}{2} + \gamma = \frac{1}{2} + \frac{k_0}{k_d} R \\
\beta &= \frac{k_a}{k_d} + \gamma = \frac{k_a}{k_d} + \frac{k_0}{k_d} R \\
\gamma &= \frac{k_0}{k_d} \frac{k_a}{k_d} R
\end{align*}
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\gamma &= \frac{k_0}{k_d} \frac{k_a}{k_d} R
\end{align*}
\]

Chemical reactions and radiolytic yields

- The chemical reactions between radiolytic species with no electrostatic interaction (i.e. their charge product is 0) can be simulated by using the Green Functions described above.
- The radiation chemistry code can be used to simulate the time evolution of the radiolytic species (radiation chemistry) and radiochemical yields [5,6].

Assumptions of the model

- The pair of particles may react as follow [3]:

\[
A + B \rightarrow (AB) \rightarrow AB
\]

\[
k_1: \text{association rate constant} \\
k_2: \text{dissociation rate constant} \\
k_3: \text{product formation rate constant}
\]

- Transitions from a state to another are defined:

<table>
<thead>
<tr>
<th>State after one timestep</th>
<th>Free (μ)</th>
<th>Rev bound (σ)</th>
<th>Product (π)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Free (μ)</td>
<td>p(0,μ)</td>
<td>p(μ,0)</td>
<td>p(1,μ)</td>
</tr>
<tr>
<td>Rev bound (σ)</td>
<td>p(0,σ)</td>
<td>p(σ,0)</td>
<td>p(1,σ)</td>
</tr>
<tr>
<td>Products (π)</td>
<td>0</td>
<td>0</td>
<td>1</td>
</tr>
</tbody>
</table>

Sampling of the Green functions

- We have developed exact algorithms to sample the random variates r for p(r, |t_r|) and p(t, |t|) [4].
- The algorithm allow the simulation to be done in several timesteps (time discretization).

Radiation track structure and evolution in time

- Time-dependent yields of chemical species produced by 300 MeV⁺ protons (LET=0.3 keVμm)[3]. Calculations: ICONYS-IRRT (---); SBS (---). The dots are experimental data.

Radiation track structure and evolution in time

- Time evolution, in 3D, of a 24-MeV⁺ "He⁺", LET=26 keVμm, at 10⁻³, 10⁻⁴, 10⁻⁵ and 10⁻⁶ s. Each dot is a radiolytic species

Many-particles system

- When more particles are added to the system, the number of interactions grow quickly
  - 2 Particles
    -1-2 (1 interaction)
  - 3 Particles
    -1-2, 1-3, 2-3 (3 interactions)
  - 4 Particles
    -1-2, 1-3, 1-4, 2-3, 2-4, 3-4 (6 interactions)
  - N Particles
    -N(N-1)/2 interactions → Grows as ~N²!

Conclusion

- This approach has been used successfully to simulate the time evolution of radiolytic species and to calculate radiochemical yields.
- The radiation track structure code RITRACKS [7] and the chemistry code will be of crucial importance in future models of DNA damage.

References