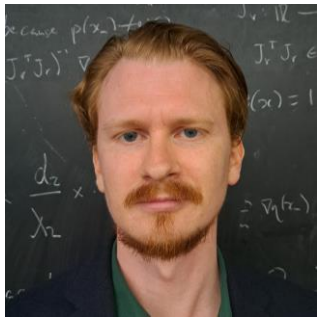
Manchester Inhomogeneous Radiation Chemistry by Linear Expansion (MIRaCLE)

Simulating Ion-beam induced chemistry: A DCF student summer project

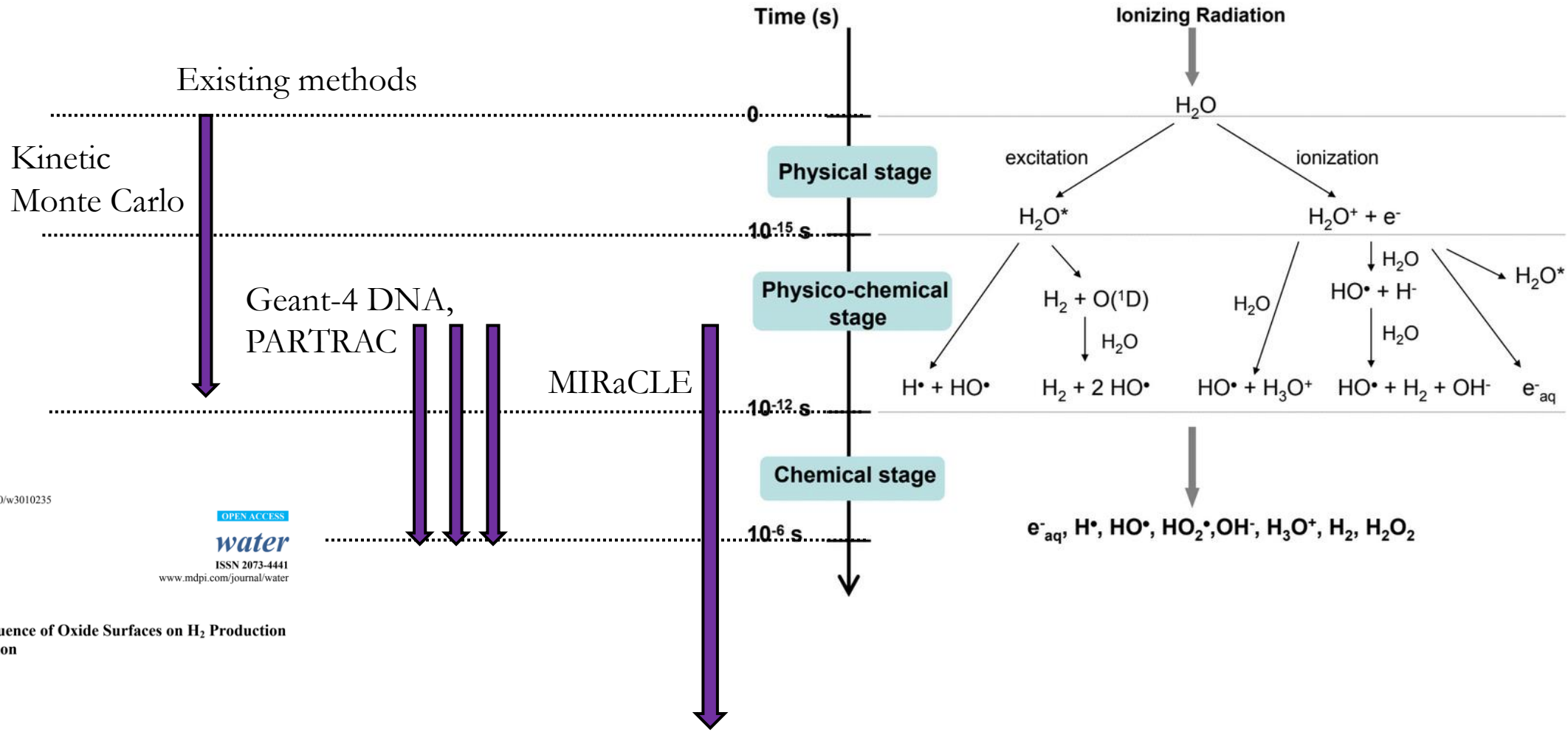
Fred Currell
Department of Chemistry



Marcus Webb
Department of Mathematics

Charlie Perkins
DCF summer student

Radiation Induced Water Chemistry



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water

ISSN 2073-4441

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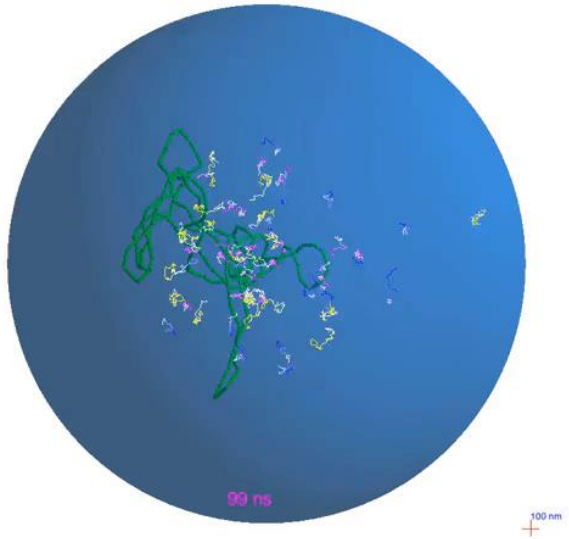
Review

Water Radiolysis: Influence of Oxide Surfaces on H_2 Production under Ionizing Radiation

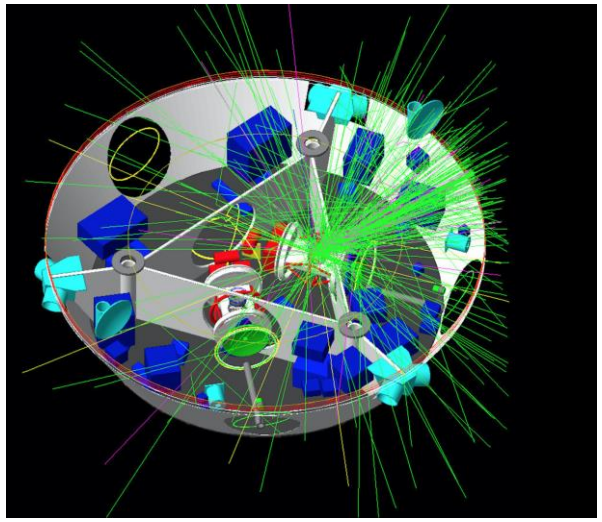
Sophie Le Ca ër

Kinetic Monte Carlo

- KMC methods keep track of each **individual species particle**.
- E.g. Geant4, PARTRAC, FLUKA, RITRACKS
- State of the art for high energies (physical stage).
- Difficulties at high densities / low energies (chemical stage).



http://geant4-dna.org/

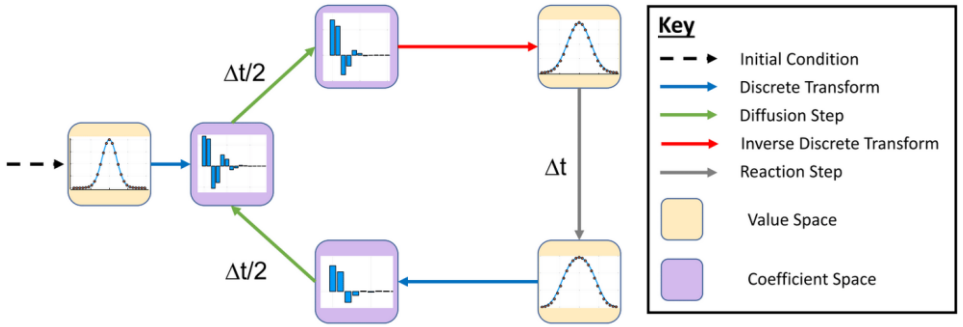


https://knowledgetransfer.web.cern.ch/technologies/geant4

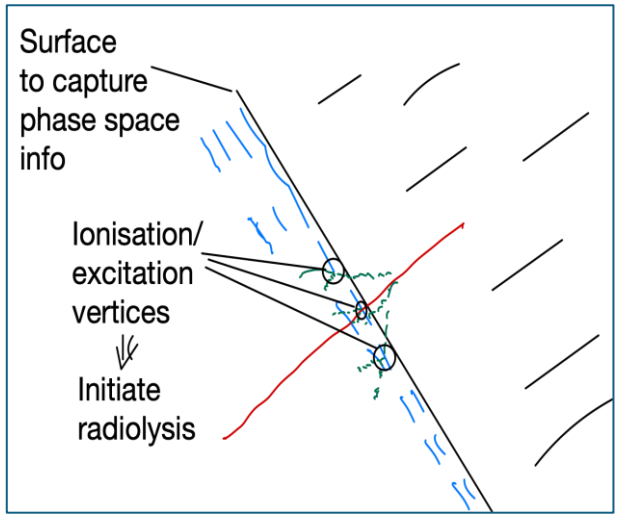
MIRaCLE Under the Bonnet

$$\frac{\partial [\mathrm{A}]}{\partial t}(\mathbf{x}, t) = \underbrace{D\,\nabla^2[\mathrm{A}](\mathbf{x}, t)}_{\text{Diffusion of A (how much does A spread?)}} + \underbrace{R_{[\mathrm{A}]}([\mathrm{A}](\mathbf{x}, t), [\mathrm{B}](\mathbf{x}, t), [\mathrm{C}](\mathbf{x}, t) \ldots),}_{\text{Reactions with A (How much A is being added or removed depending on what else, B, C, D, etc. is at } \mathbf{x} \text{ at t?)}}$$

Rate of change in concentration of 'A' at position **x** at time *t*.



An Example from the Nuclear Industry

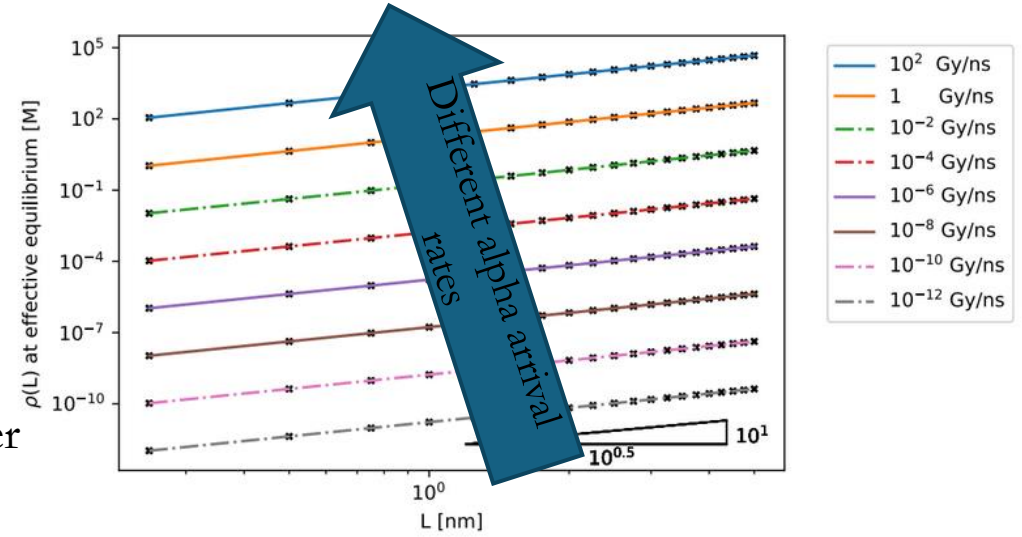


Every point on the graph corresponds to a complete dynamical simulation of a thin water layer on plutonium in storage.

Hundreds of simulations in one day on a laptop - allows us to go exploring. Previously used methods took weeks on a super-computer and still did not account for the reactive interactions with the plutonium oxide surface

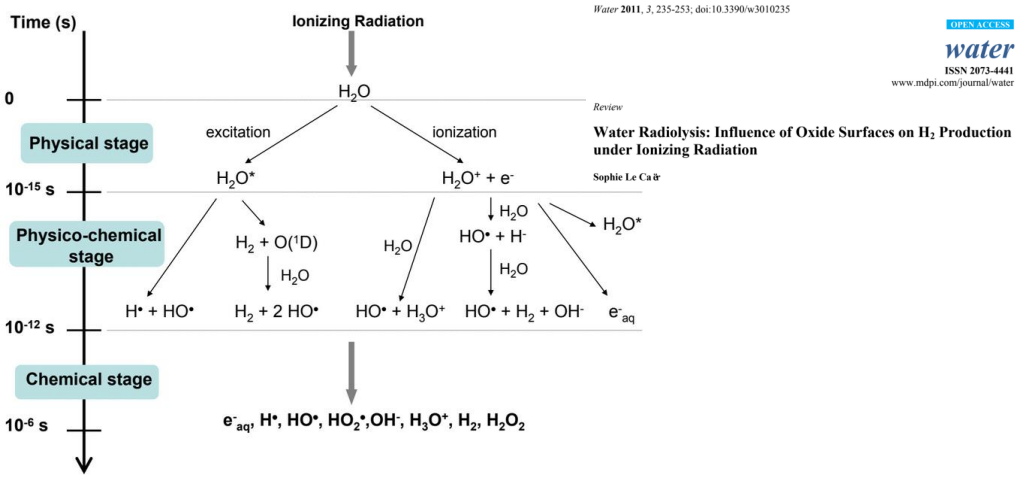
Plutonium Storage

Simulate alpha radiation interaction with thin liquid layer L
-> Find equilibrium concentration of monoatomic hydrogen
160 simulations, 14 orders of magnitude in dose rate,
15 orders of magnitude in concentration



See Bradshaw et al. 2023

My Project



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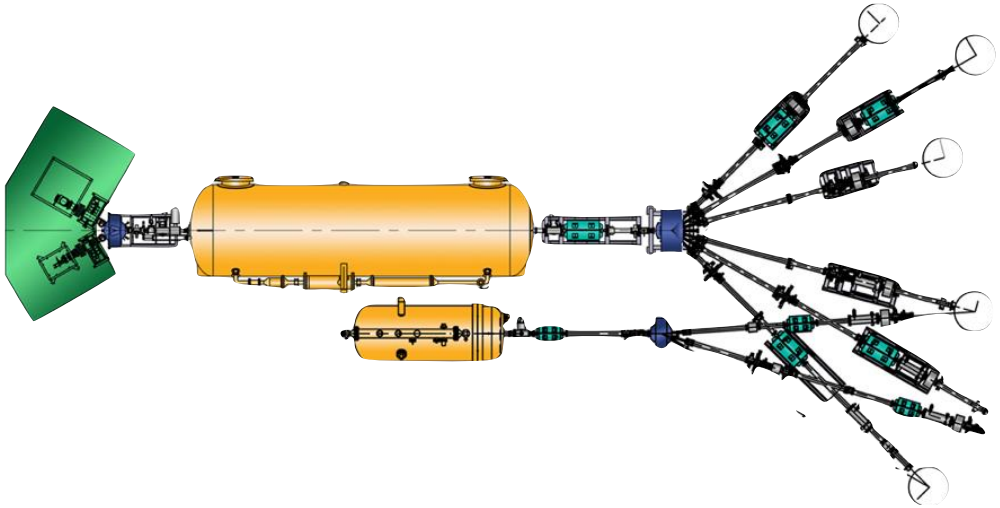
Review

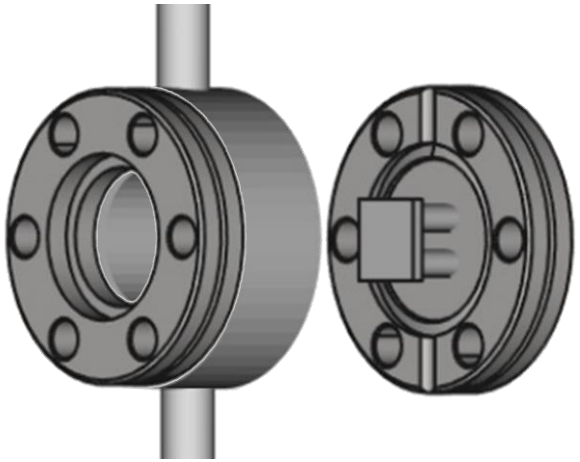
Water Radiolysis: Influence of Oxide Surfaces on H_2 Production under Ionizing Radiation

Sophie Le Ca ir

Build simple test models of relevance to DCF and beyond
Quick to simulate *and* modify

Ions into H_2O → ?





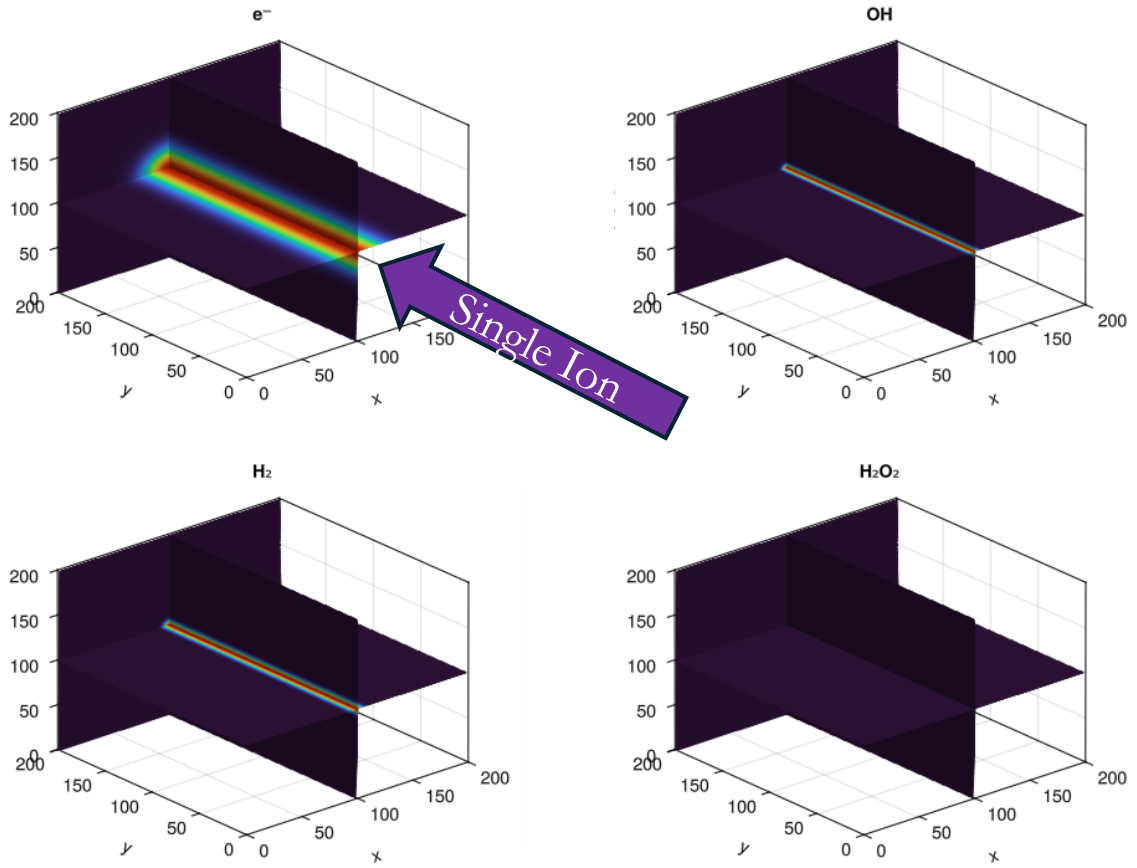
Initial Conditions & Geometry

Radiat Environ Biophys (2009) 48:11–20
DOI 10.1007/s00411-008-0194-8

ORIGINAL PAPER

Time- and space-resolved Monte Carlo study of water radiolysis for photon, electron and ion irradiation

Maximilian S. Kreipl · Werner Friedland · Herwig G. Paretzke

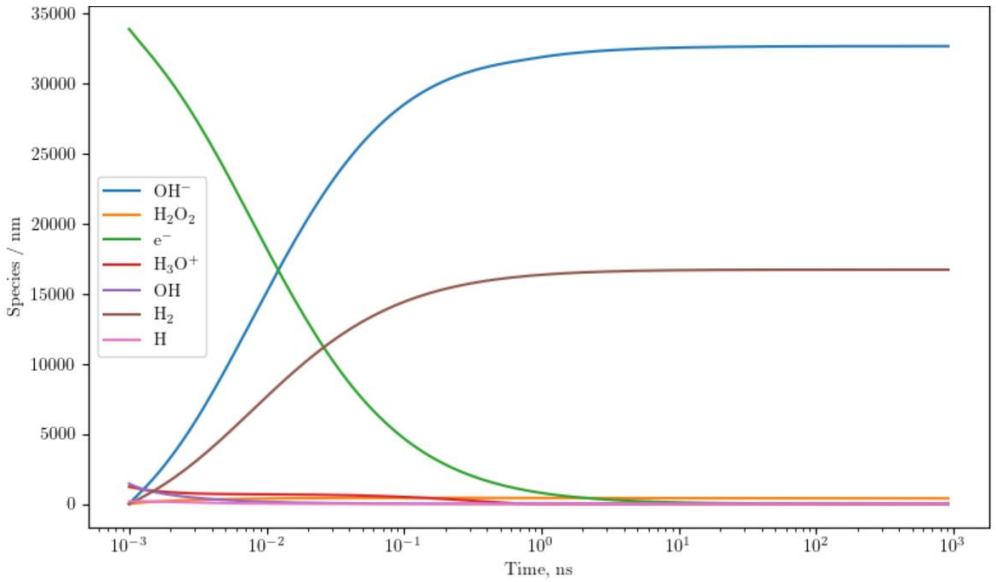


Treat single ion traversal
G-values from Kreipl paper
Initial concentrations from physicochemical stage + LET
Reduced to 1D (faster to simulate)

Simulation of Time Dependence

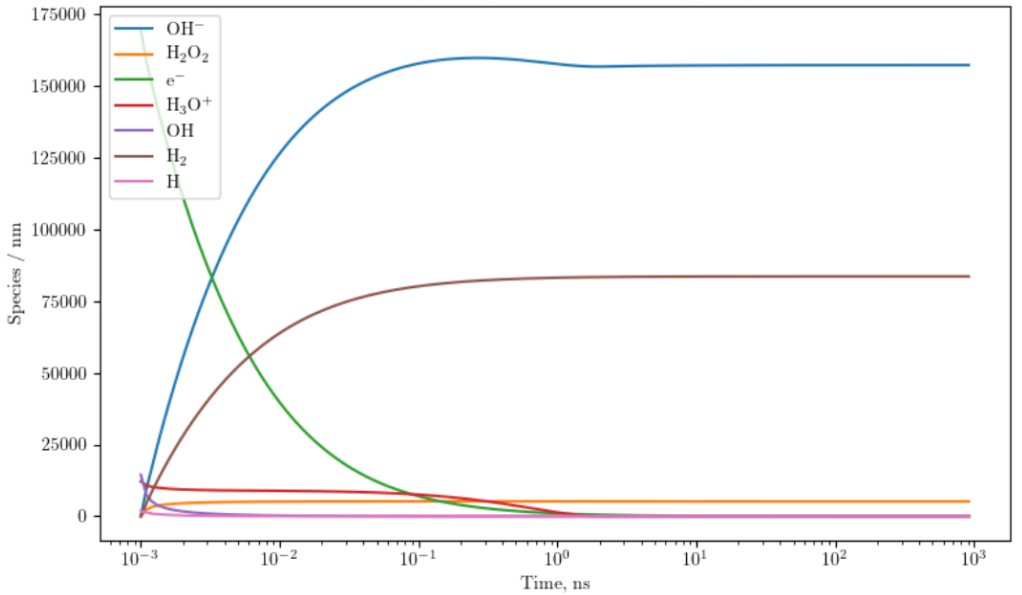
- Initial Conditions: Average effect of radiation on H_2O over different ions
- Easily trial different LETs
- Very fast simulation times

Single Ion, LET 10keV/µm , e.g. ~1 MeV $^1H^+$



Simulation time 1s

Single Ion, LET 100keV/µm , e.g. 75 keV $^4He^{2+}$

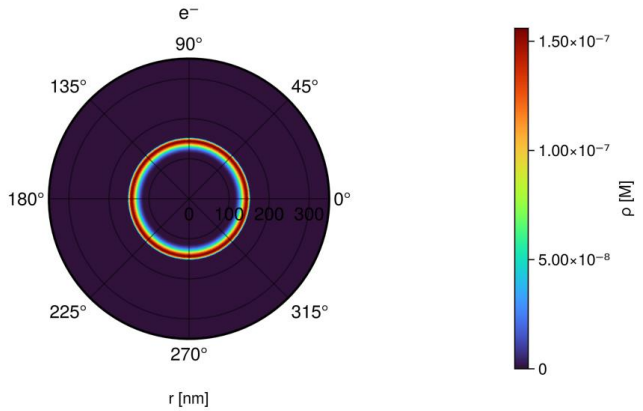


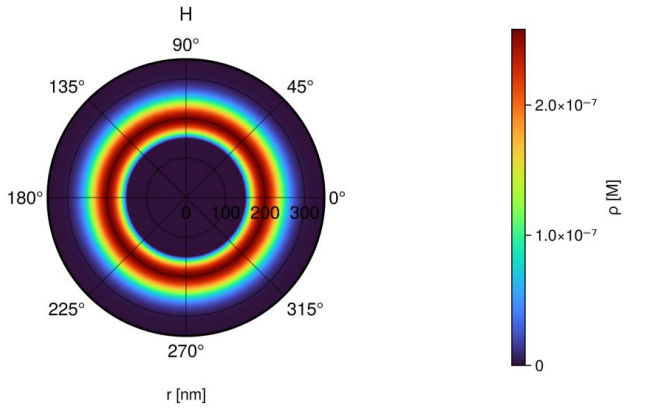
Simulation time 2s

A Holey MIRaCLE

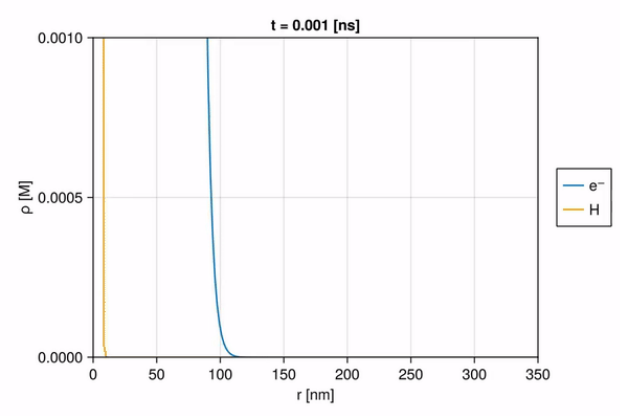
Single Ion, LET 10keV/μm , e.g. ~1 MeV Proton

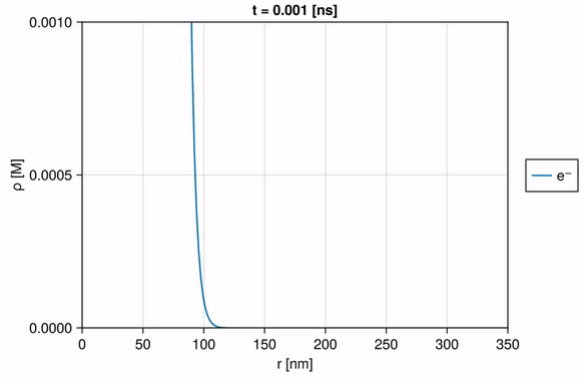
e^-_{aq}, H distribution hollowing out (500ns)



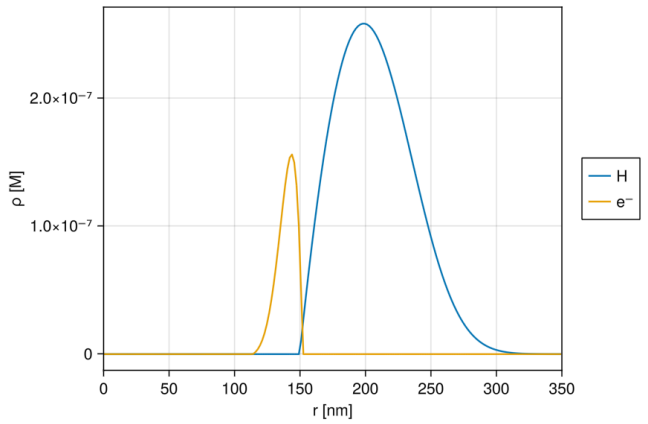


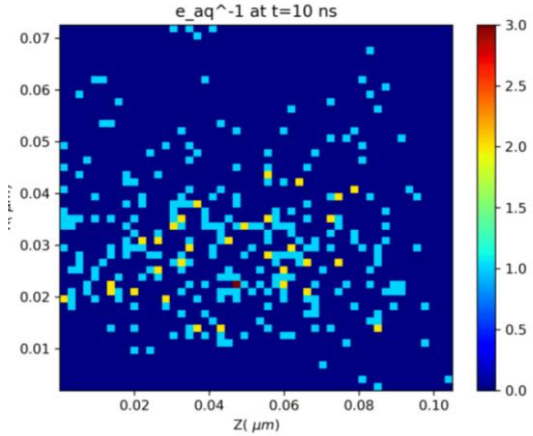
$$H^\bullet + e^-_{aq} + H_2O \rightarrow OH^- + H_2$$





Hollowing at 500ns





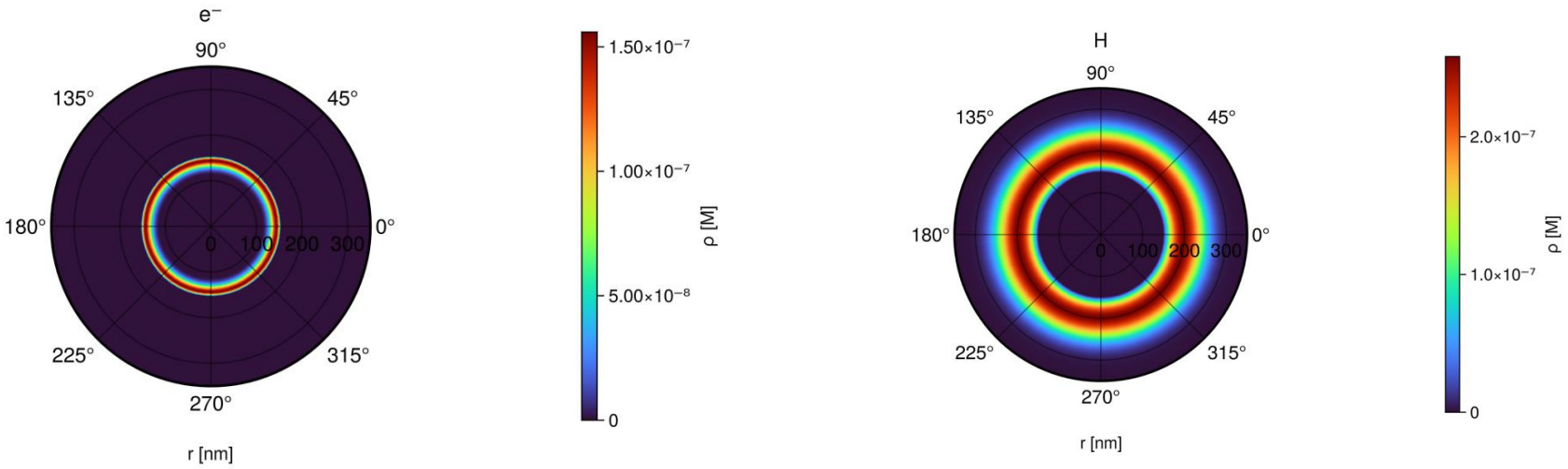
Monte Carlo:
Balder Villagomez

Conclusions

Interesting phenomena emerges from MIRaCLE simulations of water radiolysis
MIRaCLE is a fast, easy to use multipurpose tool for solving the reaction-diffusion equation
MIRaCLE provides an easy basis for DCF users to quickly simulate complex reactions

Other possible applications of MIRaCLE:

- **Not just water!**
- Atmospheric science
- Planetary science
- Civil nuclear
- Material damage
- …



Aside: Particles v Continuum (Tran et al. 2021)

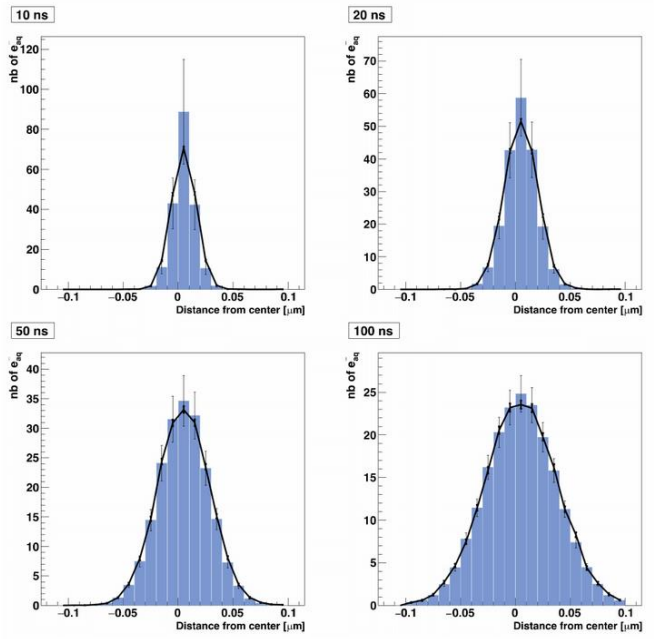
International Journal of Molecular Sciences
MDPI

Article

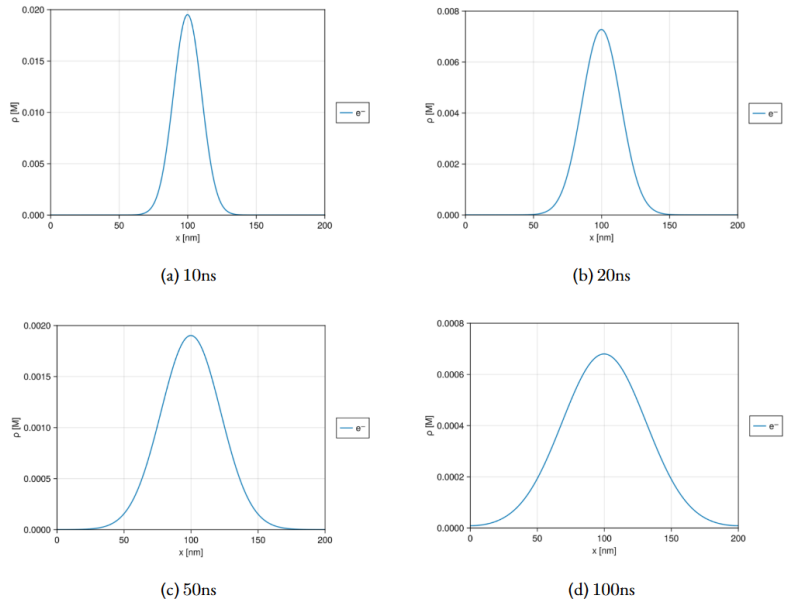
Geant4-DNA Modeling of Water Radiolysis beyond the Microsecond: An On-Lattice Stochastic Approach

Hoang Ngoc Tran [1,*], Flore Chappuis [2], Sébastien Incerti [1], Francois Bochud [2] and Laurent Desorgher [2]

Tran Pure Diffusion



MIRaCLE Pure Diffusion



(a) 10ns (b) 20ns

(c) 50ns (d) 100ns

MIRaCLE performs all diffusion steps analytically
-> Only error is due to initial state approximation

Aside: Particles v Continuum (Tran et al. 2021)

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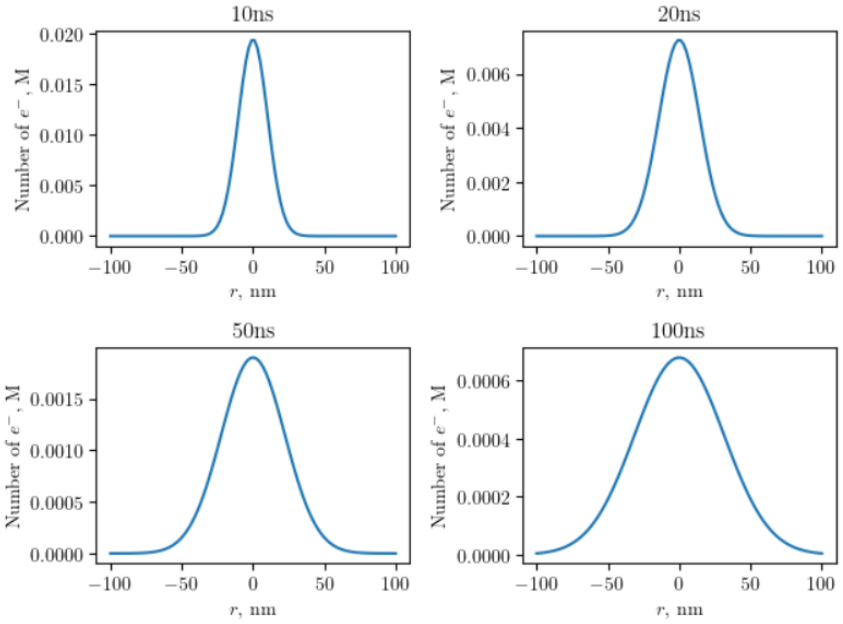
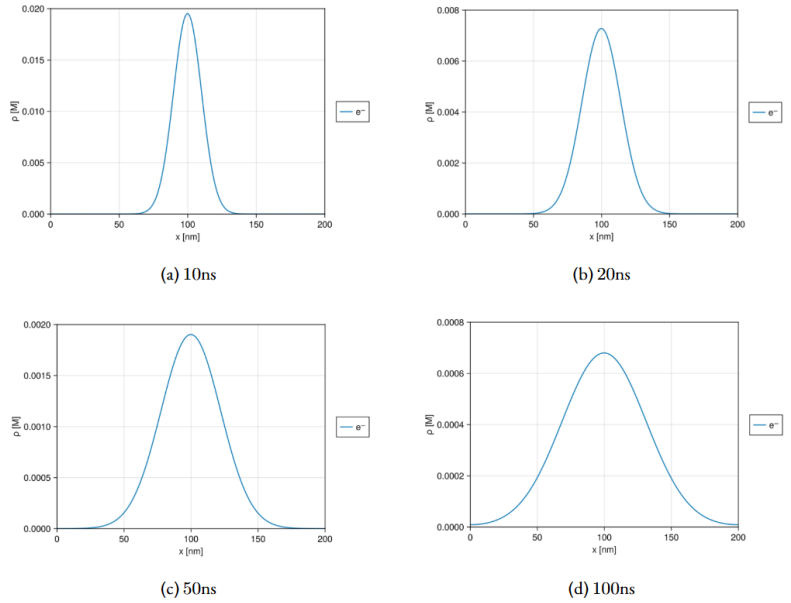
Analytic Diffusion of Top Hat



MIRaCLE Pure Diffusion



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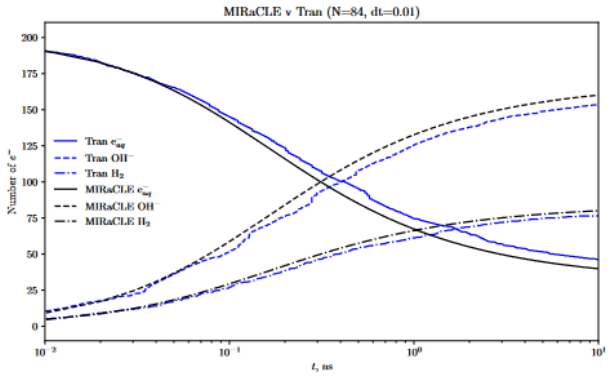
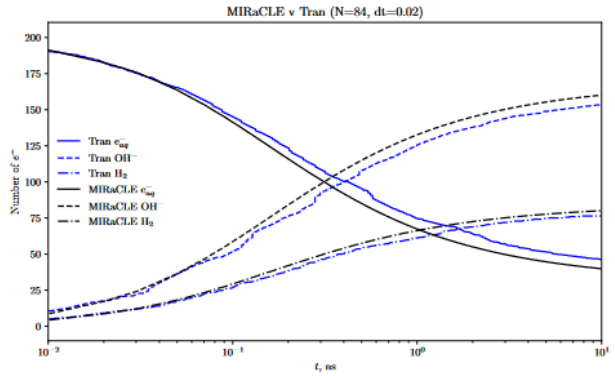
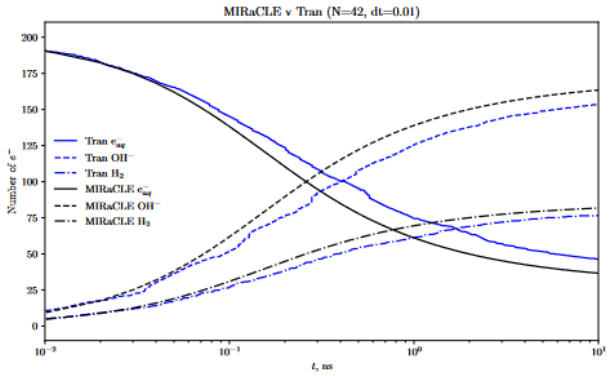


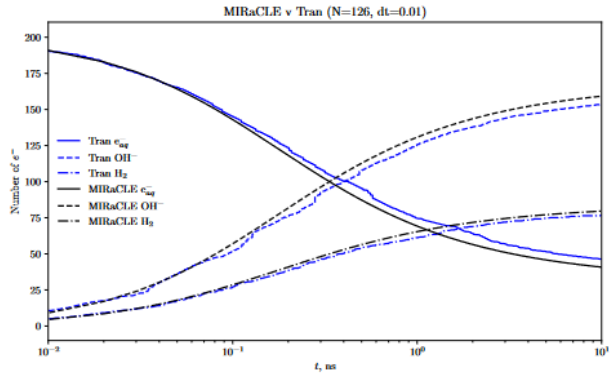
(a) Home



(b) $dt \rightarrow dt \times 2$



(c) $N \rightarrow N/2$



(d) $N \rightarrow N \times 3/2$

International Journal of *Molecular Sciences*

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Simple reaction scheme
Tran (Blue)
MIRaCLE (Black)