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# Radiation Chemistry Experiments at Upgraded CLARA Facility

Alex Baidak, The University of Manchester

4th CLARA User Meeting, 03-Dec-2024

NNUF National Nuclear User Facility

HENRY ROYCE INSTITUTE

UKNIBC UK NATIONAL ION BEAM CENTRE

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# The Dalton Cumbrian Facility (DCF)


**Purpose:** foster and support the understanding of radiation-driven processes and the responsible use of radiation.
**Vision:** support innovation in applications of radiation science and engineering.

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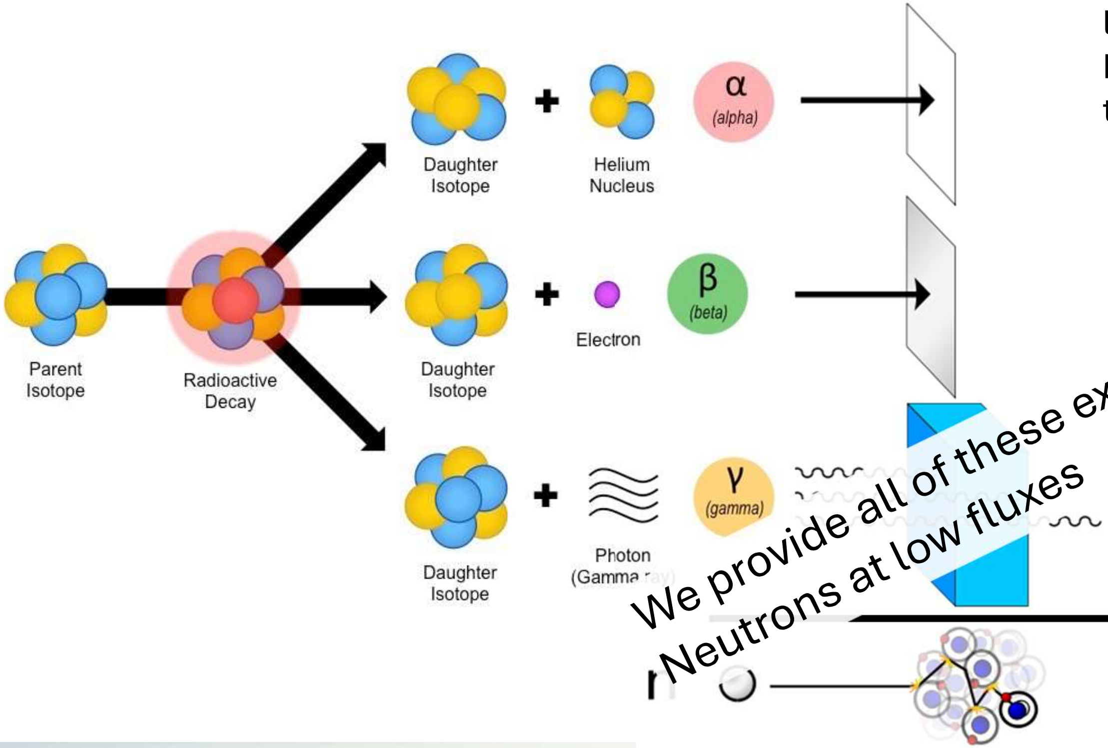
# DCF's Radiation Provision




We provide all of these except betas,
Neutrons at low fluxes

low penetration/
high linear energy
transfer

high penetration/
low linear energy
transfer

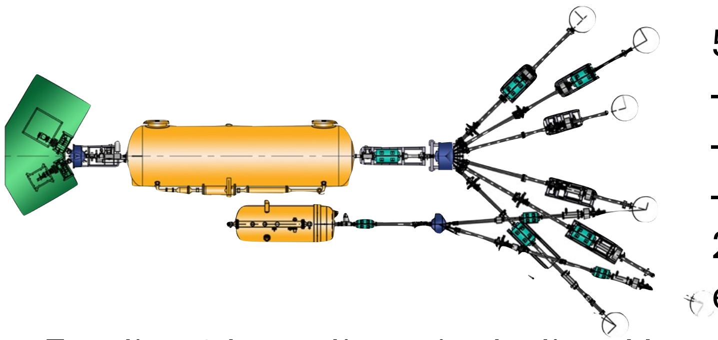
low displacement
per atom (DPA)

p

Heavier ions too!

high displacement
per atom (DPA)

# DCF's Radiation Sources



**Two ion accelerators**
5MV tandem ion accelerator
–10 MeV $^{1}H^{+}$ up to 100 μA,
–15 MeV $^{4}He^{2+}$ up to 15 μA
–35 MeV $^{12}C^{6+}$ up to 150 nA
2.5MV single ended ion accelerator
e.g. $^{1}H^{+}$ and $^{4}He^{2+}$

Feeding 6 beamlines, including: Hot cell (active samples) Dual Beam, Extensive Characterisation (SIMS, EELS, Tensile Rig, ...)

**Foss Therapy Services**
**812 $^{60}Co$ irradiator**

~ 1.2 MeV gamma rays
~ 330 Gy/min – 30 Gy/hr

**X-ray cabinet source**
**Precision MultiRad350**

30 – 350 kVp X-rays
up to 140 Gy/min

**All supported by:**

- A team of 12 technical specialists
- Extensive on-site analytical capabilities
- Robotic/automated sample handling
- Cutting-edge radiation transport and radiation chemistry modelling

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# Why The Dalton Cumbrian Facility?
# Because Radiation Changes Matter

## Energy

**Legacy**

Reprocessing

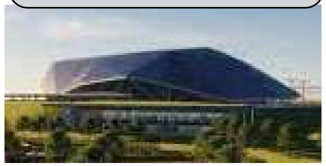
e.g. plutonium stockpile

Storage

e.g. UK GDF

**Future**

Fission



e.g. SMR, new designs

Fusion

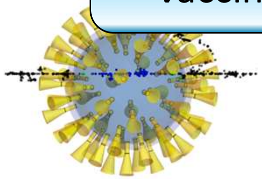
e.g. STEP

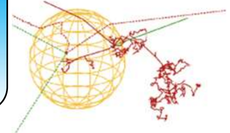
## Advanced Materials

- Polymers and composites
- Nanomaterials
- New dosimeters

## Health

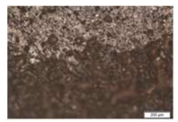
- Rapid vaccines
- Nuclear medicines
- Advanced cancer therapies





## Space

- Space hardware
- Lunar base materials



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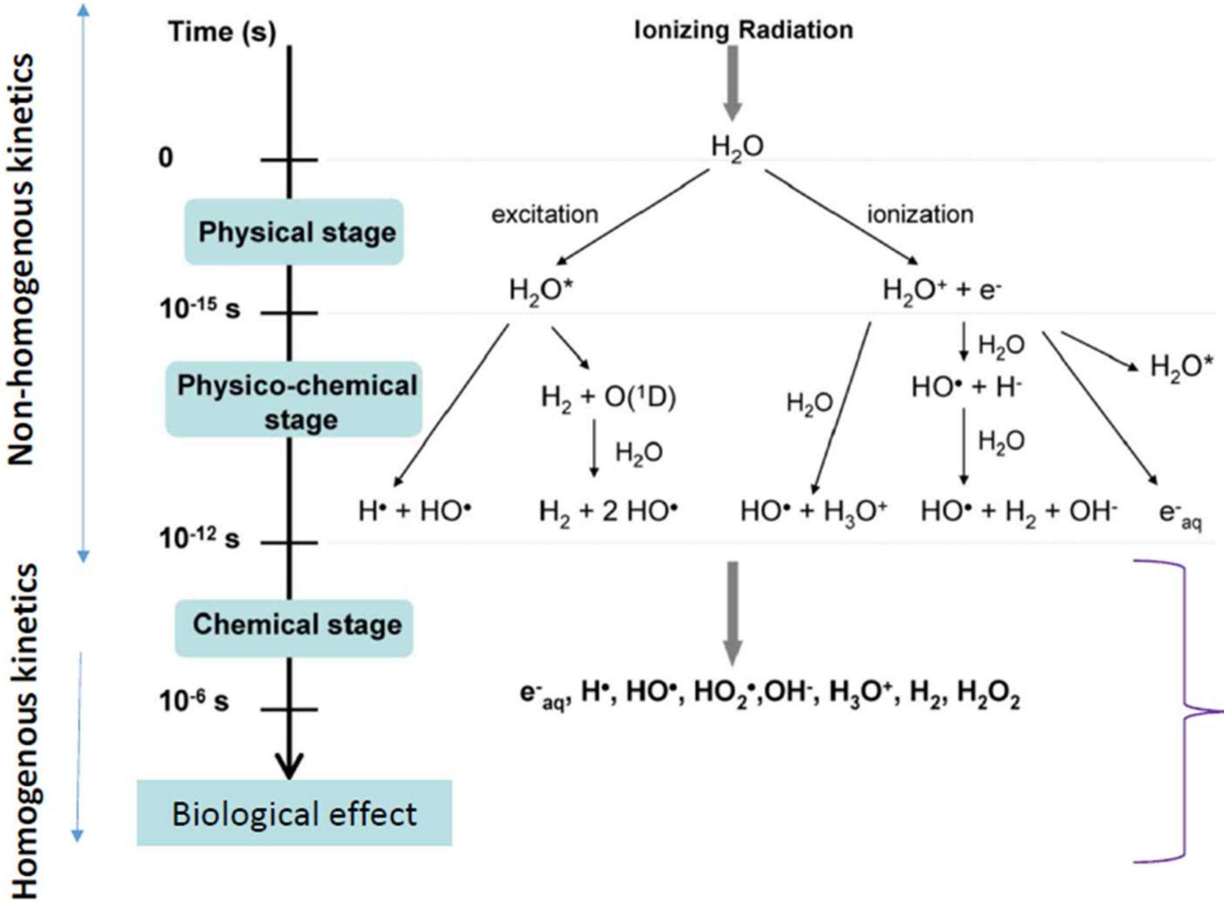
# Interaction of Radiation with Water




*Water* **2011**, 3(1), 235-253; doi:10.3390/w3010235
Open Access
*Review*

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

Sophie Le Caër

Novel science,
Domain of discovery

Useful kinetic information,
well studied for electrons,
still novel for ions

# Unresolved Questions in Ultrafast Radiation Chemistry

**Primary ionization step:** $H_2O \longrightarrow H_2O^+ + e^-$

**Elementary proton transfer:** $H_2O^+ + H_2O \longrightarrow OH + H_3O^+$

- Lifetime of the radical cation as a function of T and pH
- Absorption spectra of the $H_2O^+$ and ·OH under different conditions
- The extent of delocalisation of the hole in $H_2O^+$
- Dynamics of the solvent rearrangement upon ionisation
- How the electron dynamics couples to the ion dynamics
- Pre-solvated electron capture
- Dissociative electron capture

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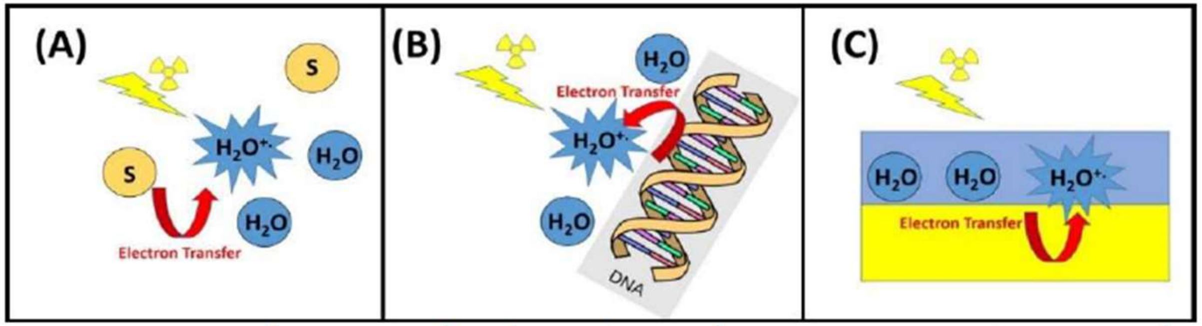
# Ultrafast Radiation Chemistry: Possible Scenarios




Radiolysed water can form radical cation that engages in an electron transfer in competition with the proton transfer:

(A) highly concentrated solutions in which the water radical cation can oxidise solute molecules directly – e.g. spent nuclear fuel processing (PUREX)

(B) highly-structured water layers formed in contact with biomolecules, such as DNA – cancer therapy

(C) water/solid interface – safe nuclear waste storage, Pu stewardship, corrosion in reactors and storage ponds.

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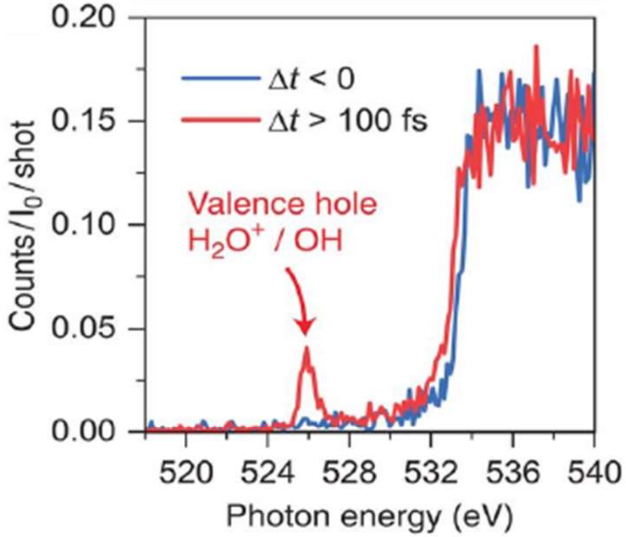
# Potential Experiment 1: Water Radiolysis under Extreme pH

**Pure water:**

Observing proton transfer via OH radical signature

$$H_2O^+ + H_2O \rightarrow OH + H_3O^+$$

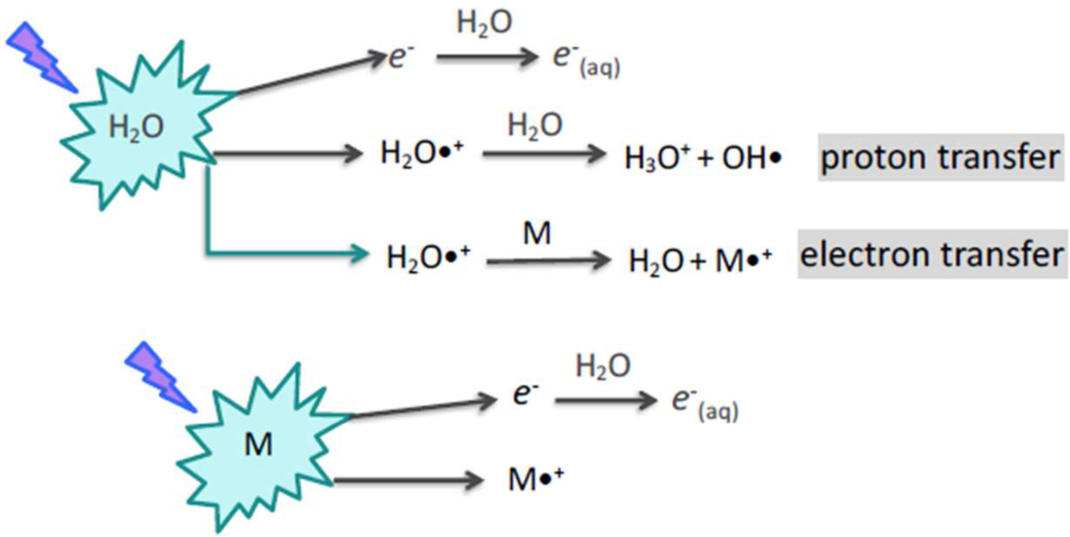



- Clean hydroxyl radical signature in water window
- Used as a tracer for reaction rates

*Science* 367, 179–182 (2020)

**Highly alkaline system:**

Competition between proton and electron transfer




- New reactants formed in spur if M is a nearest neighbor

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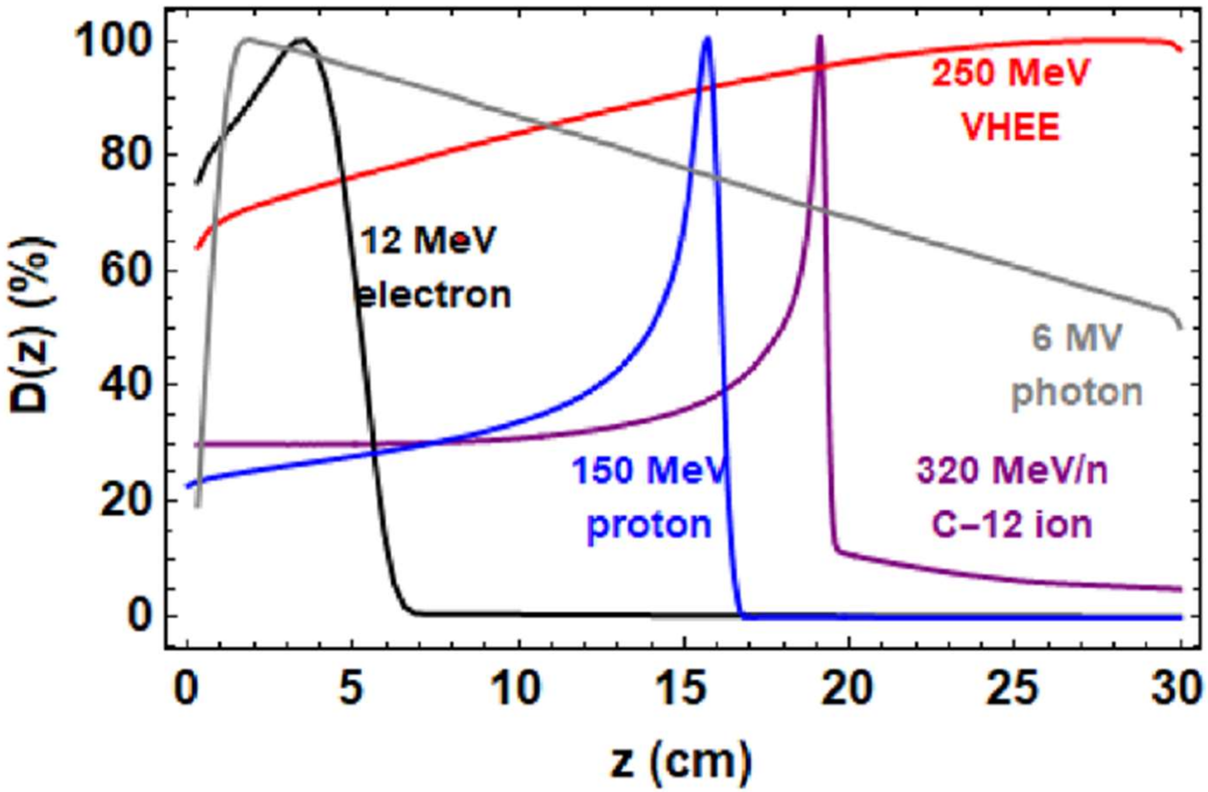
# Potential Experiment 2: Radiation Damage to DNA/RNA




Scientific Reports | (2021) 11:14013 | https://doi.org/10.1038/s41598-021-93276-8

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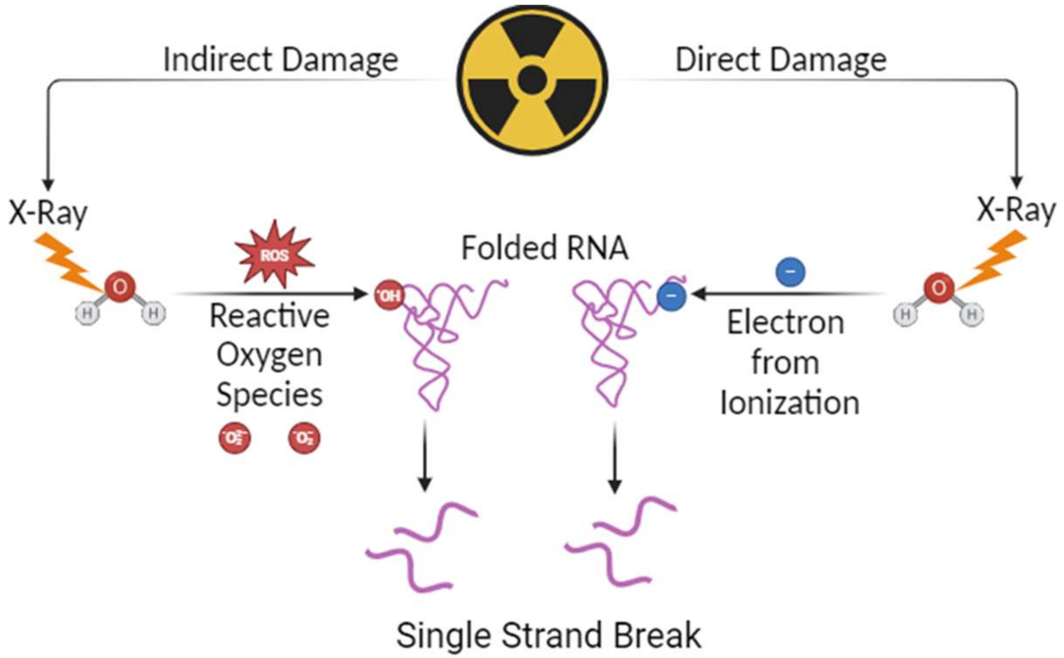
# Ionising radiation has two main damage pathways



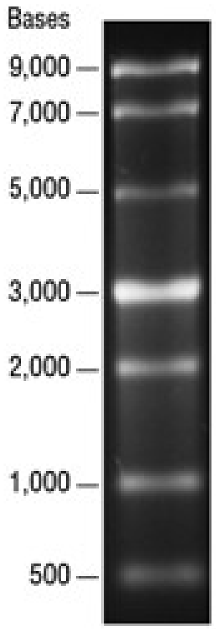

$$H_2O \xrightarrow{\text{Ionizing Radiation}} e^-_{aq}, H\cdot, HO\cdot, HO_2\cdot, OH^-, H_3O^+, H_2, H_2O_2$$

Equation 1: Water Radiolysis

Method

# RNA Sample

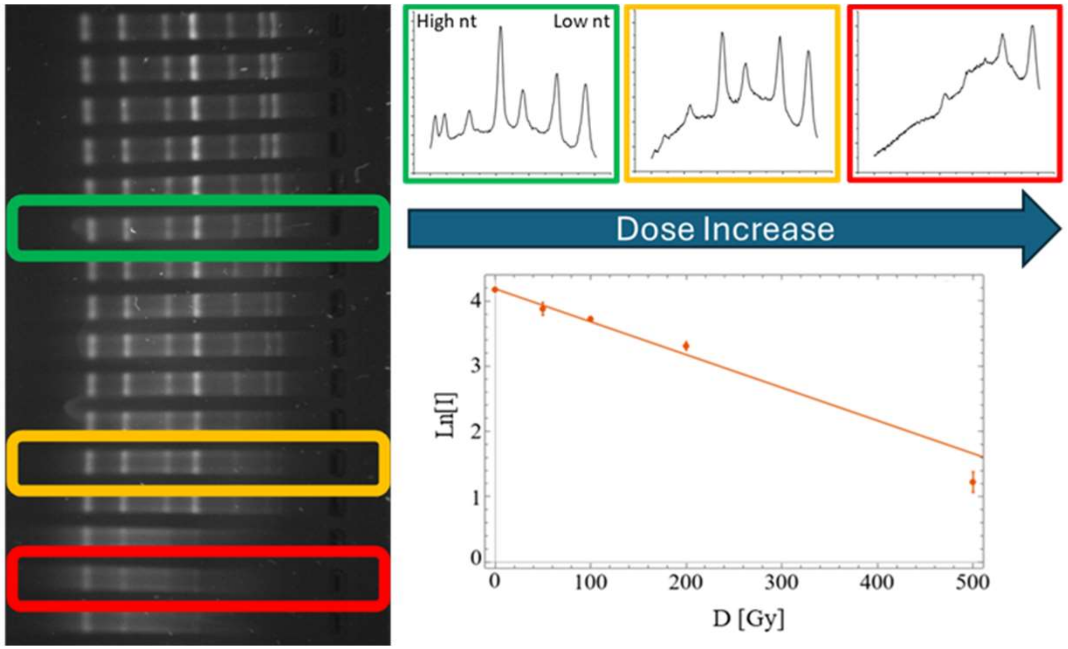
- Size range- 500 bases to 9,000 bases.
  - 9000, 7000, 5000, 3000, 2000, 1000 & 500.
- 3000 base fragment is twice as intense to serve as reference.
- Storage buffer:
  - 20 mM sodium citrate, 1 mM EDTA, pH 6 at 25°C.




Single stranded RNA Ladder

* Ongoing PhD project at the University of Manchester (PhD candidate - Jordan Elliot)

# Method Outline




- The RNA and buffers are prepared individually and then mixed.
- The RNA samples are the irradiated:

- After irradiation, the RNA samples are collected and prepared for gel electrophoresis.
- The images collected from gel electrophoresis are analysed via ImageJ and a bespoke Mathematica program.

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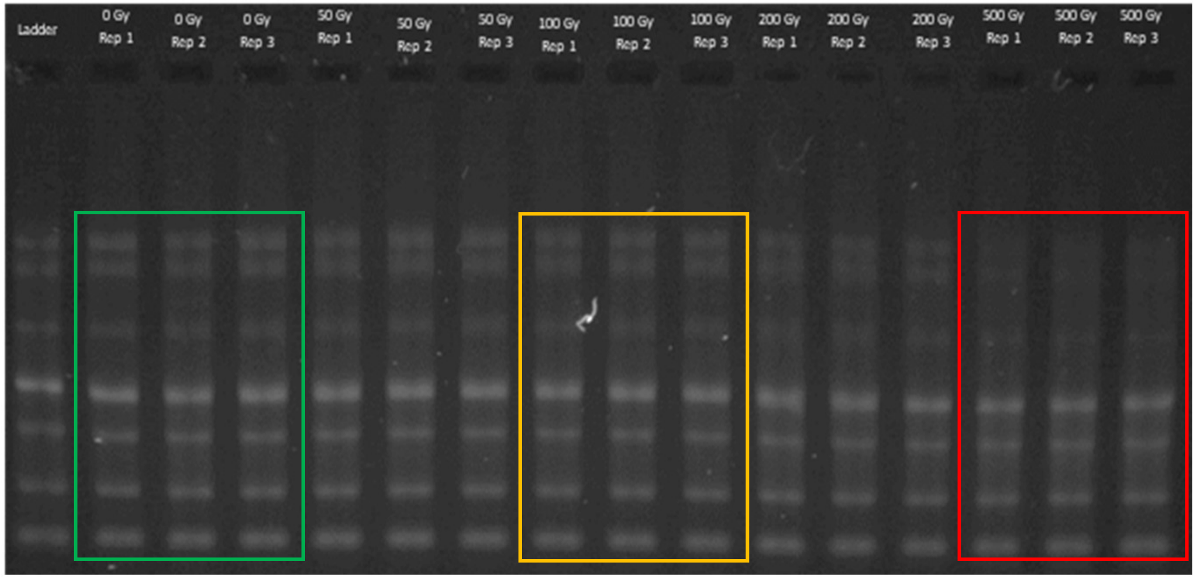
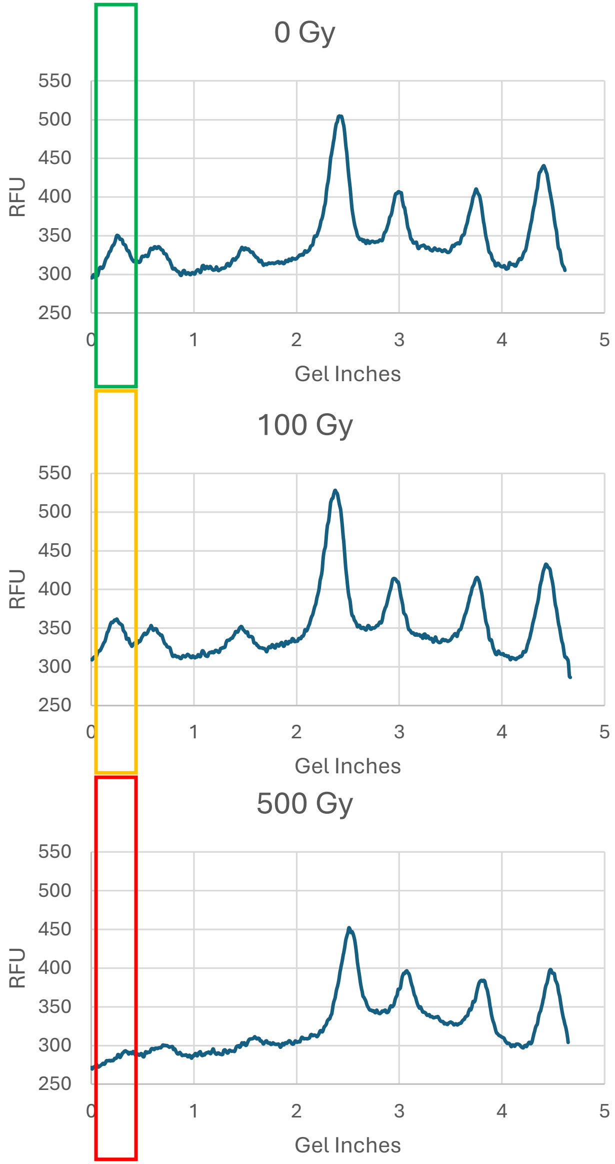
# Gel analysis

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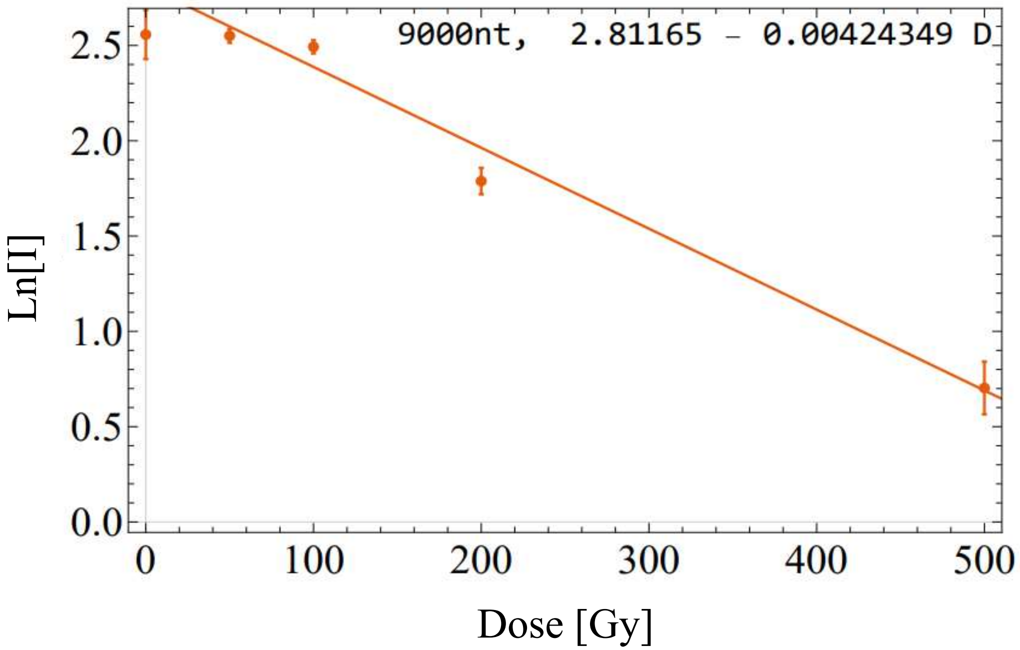
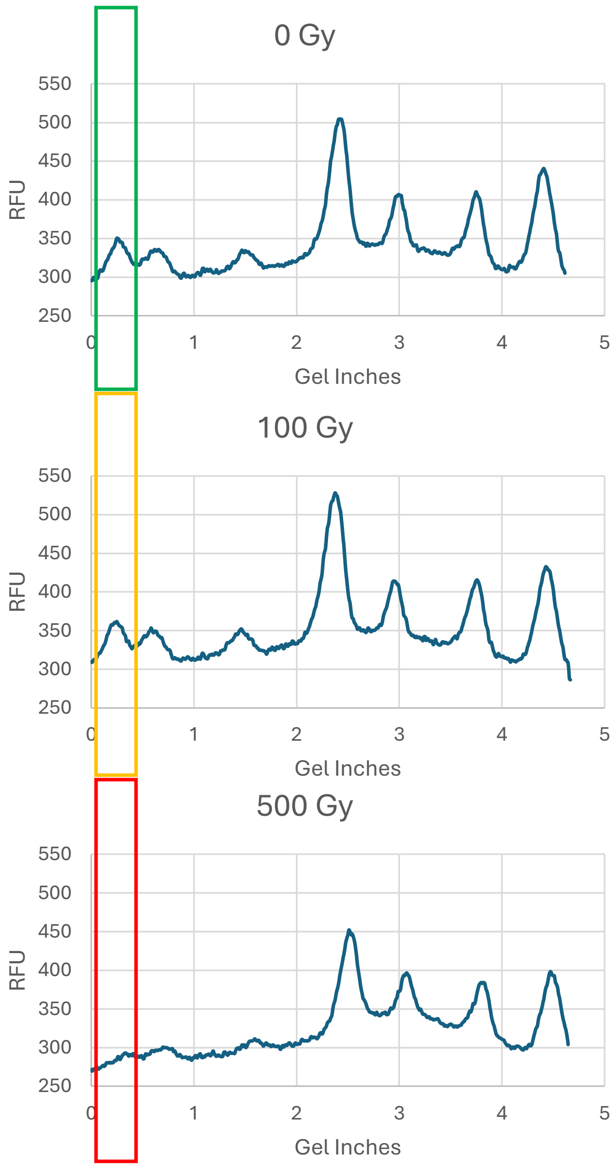
Method

# Gel analysis

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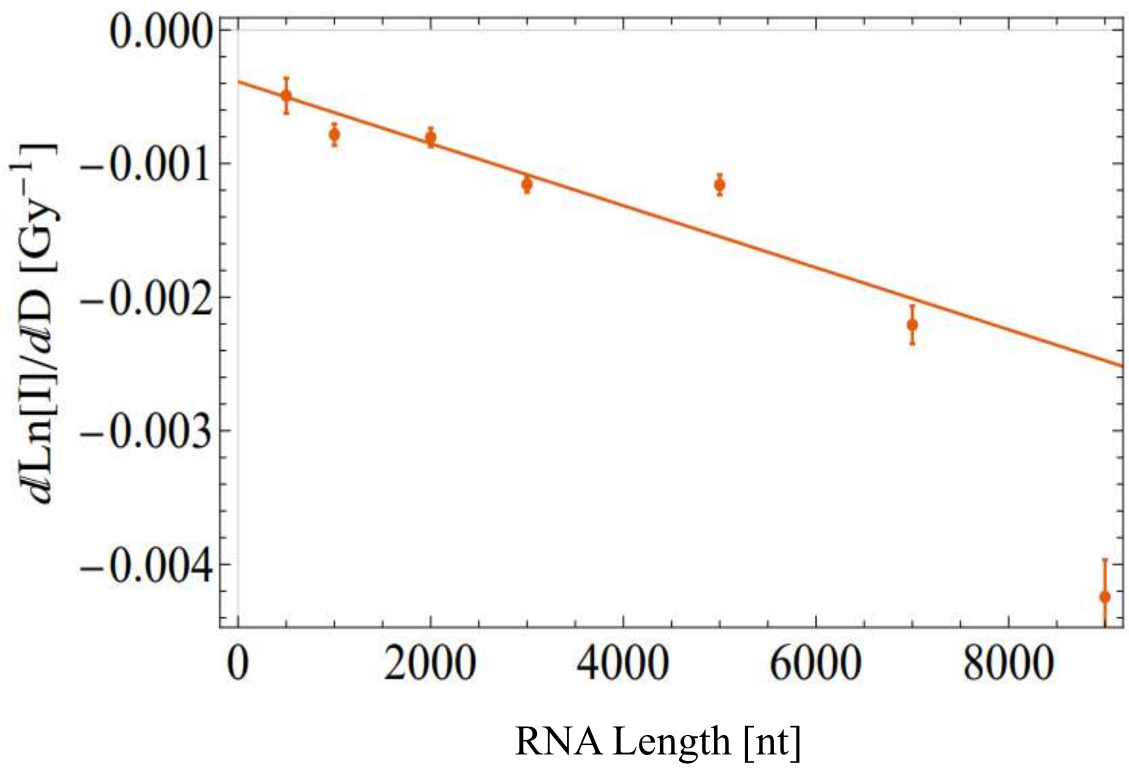
Results

# Gel analysis

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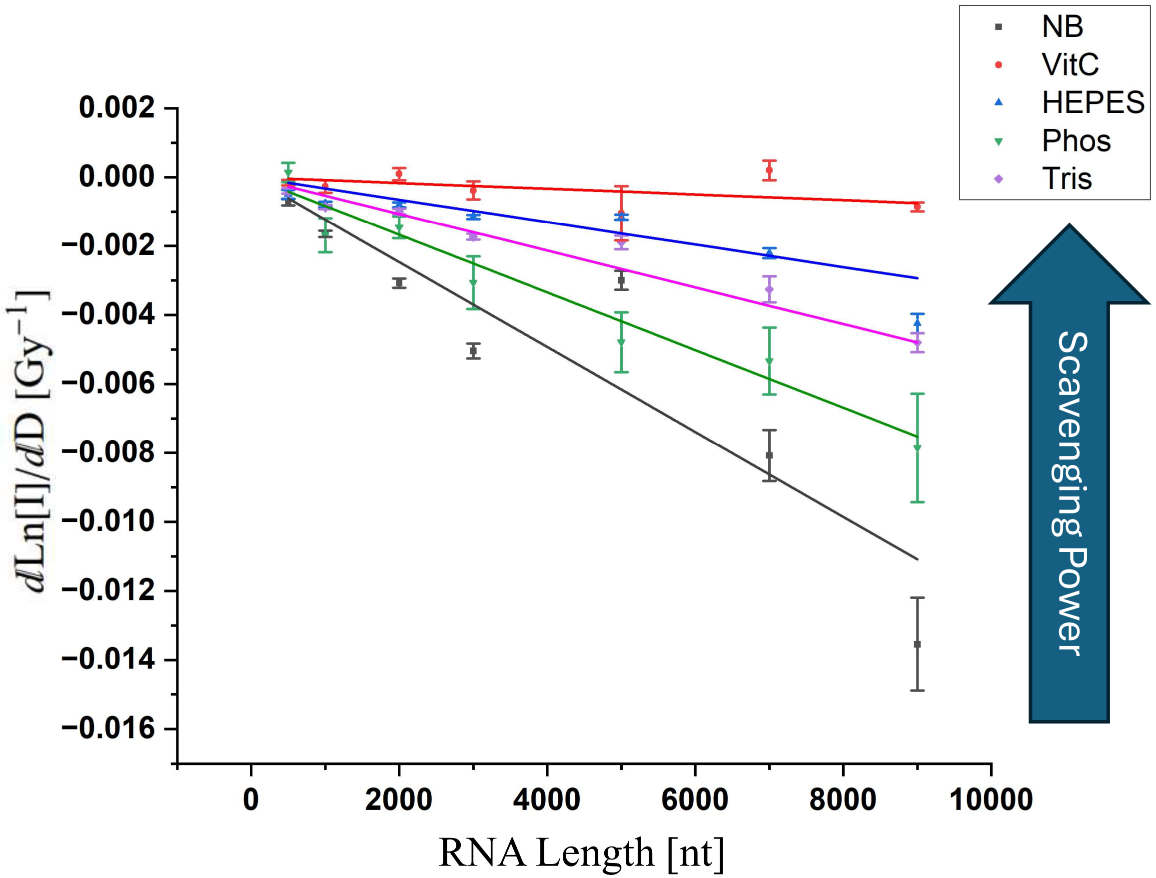
Results

# Comparison of RNA damage in different scavenger buffers

| Buffer | Rate constant for OH· radicals (L mol$^{-1}$s$^{-1}$) |
|---|---|
| Ascorbic Acid | $1 \times 10^{10}$ |
| HEPES | $6.9 \times 10^{9}$ |
| Tris | $1.5 \times 10^{9}$ |
| Phosphate | $<1 \times 10^{7}$ |

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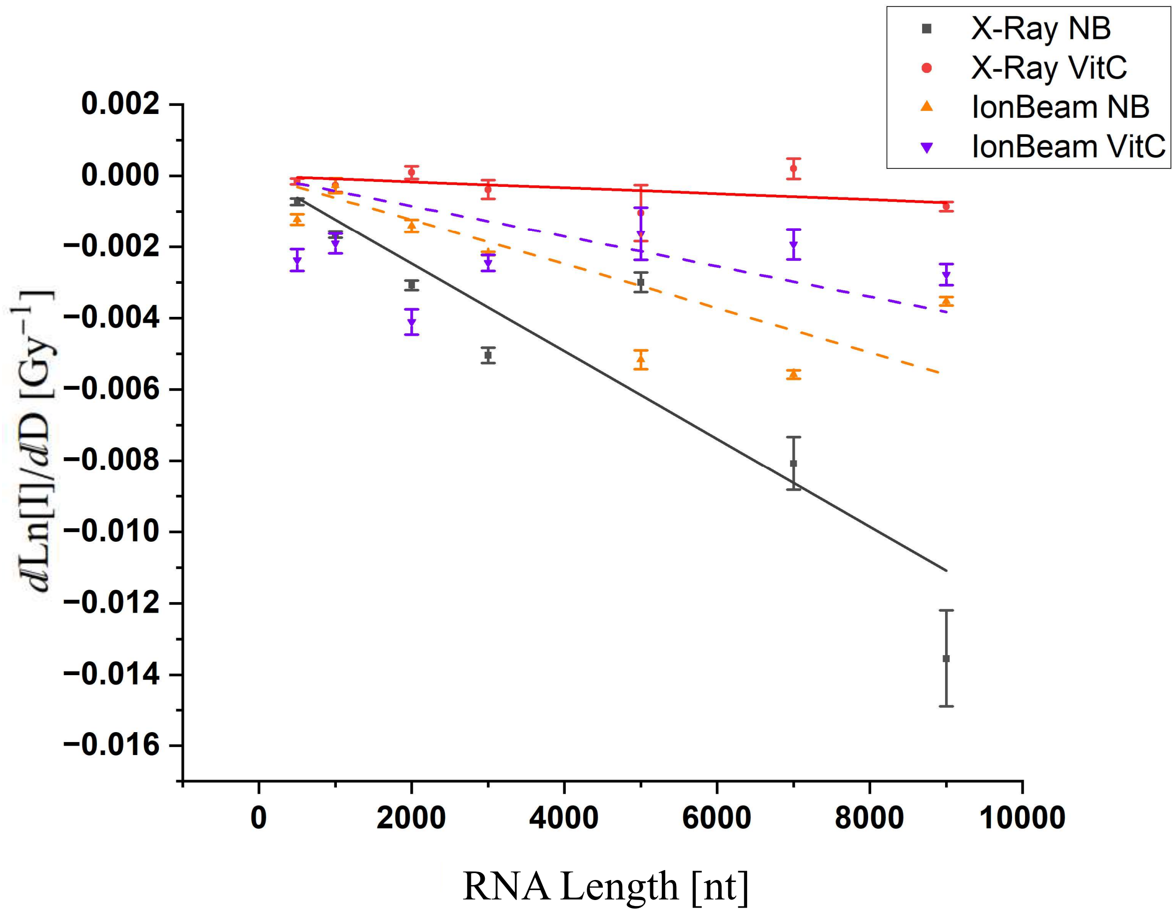
Results

# Comparison of RNA Damage: X-Ray vs Ion Beam

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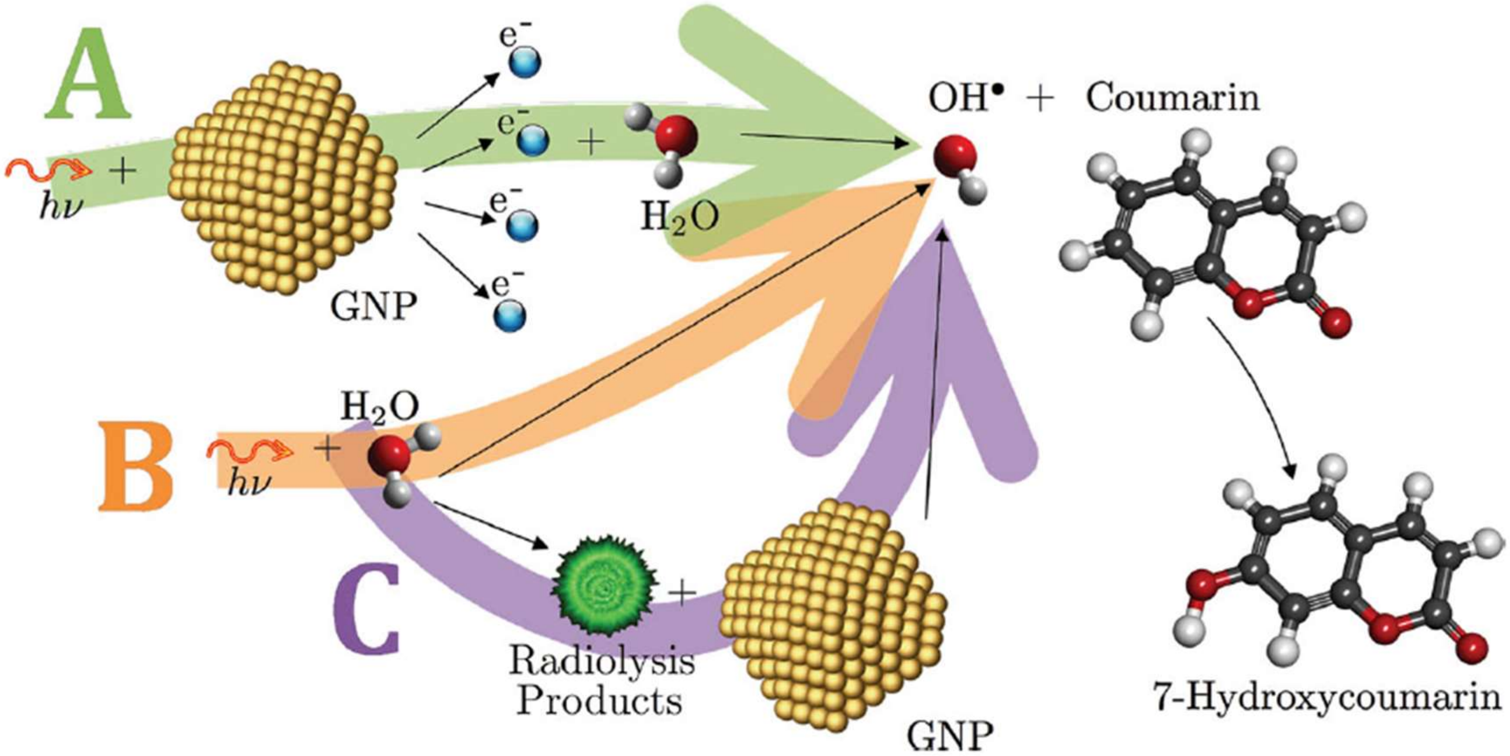
# Open Questions

- How do VHEE damage RNA/DNA in comparison to photons, ions and lower energy electrons?
- How does the efficiency of antioxidant protection against VHEE induced damage compares to that of other radiation types?
- Dose rate effects?
- Oxygen depletion during irradiation.

# Potential Experiment 3: Radiolytic Processes on Nanoparticle Interfaces




small 2014, *10*, No. 16, 3338–3346

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# Summary

- Radiolysis is complex, interesting and longstanding problem;
- Ultrafast experiments will allow to observe and control interspur dynamics;
- Insights into mechanisms in solution and at interfaces;
- Three potential radiation chemistry experiments proposed:

(a) Water radiolysis under extreme pH (strongly alkaline vs. acidic); requires time-resolved detection

(b) Radiation damage to DNA/RNA and evaluation of antioxidant protection; doesn't need time-resolved detection

(c) Radiation effects on NP interfaces; doesn't need time-resolved detection but could benefit from it.