Review of RF photoinjector for radiation chemistry

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Application for ultra-short pulse
— Radiation Chemistry experiments

**Purpose of the sub-ps pulse radiolysis**
- Investigation of the elementary process of radiation induced phenomena which occur in the time scale of ps, even sub-ps
Application for ultra-short pulse Radiation Chemistry experiments

Time scale

< asec

Excitation

H₂O

Ionization

e⁻ + H₂O⁺

Final target!

≈ fs

H + OH (H₂ + O)

OH + H₃O⁺

eₐq⁻

≈ ps

eₐq⁻, H₃O⁺, H, OH, (H₂)

≈ μs

eₐq⁻, H₃O⁺, H, OH, H₂, H₂O₂, (HO₂)

<table>
<thead>
<tr>
<th>0</th>
<th>1fs</th>
<th>1ps</th>
<th>1μs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Physical stage</td>
<td>Physicochemical stage</td>
<td>Chemical stage</td>
<td></td>
</tr>
<tr>
<td>Energy deposition</td>
<td>Ionization &amp; Excitation</td>
<td>Inhomogeneous spur reactions - diffusion &amp; reaction control</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Recombination</td>
<td>Thermalization Reorganization</td>
<td></td>
</tr>
</tbody>
</table>
Radiation Chemistry

Pulse radiolysis method

Chemical reaction of water

NERS U. Tokyo Y. Muroya et al.,
Requirements

*Pulse radiolysis in a time range of sub-picosecond*

I  Ultra-short bunch and laser

II  Stable synchronization

III  Intense electron bunch

For Pumping beam
- Short pulse
- Single beam, low dark current
- High intensity

For Probe beam
- Short pulse
- Synchronization to pumping beam
- Tunable wavelength

fs laser (Ti:Sapphire laser) + Photocathode RF gun

Suitable combination
Precise Synchronization System at UTNS

18L Linac

Laser photocathode RF gun

Compressor

THG

Accelerating Tube

RF

Compressor

50% BS

Chicane

RF gun

Klystron

15MW

Regenerative Amplifier

with Pulse Selector

Multi-pass Amplifier

Stretcher

Mode-Locker

Temperature control within 0.1 deg Clean room (class : 10,000)

Digitex

To Streak Camera

To Pulse Selector

Master Oscillator

119MHz

x 4

x 6

3DB

x 1/5

x 1/6

50Hz

Trigger Pulse

Timing Stabilizer at 9th Harmonics

Diode Pump Laser

Ti:Sapphire Laser System

Femtosecond Ti: Sapphire Oscillator

with Kerr Lens

Mode-Locker

Cherenkov Radiator

Femtosecond Streak Camera

Laser transport line
Measurement system

Laser (265nm) → Linac → Sample

PD1, PD2

Shutter1 (On/Off)

FC

Laser (265nm)

Shutter2 (On/Off)

Delay stage

Fs laser (795nm)

BPF(400-1100nm)

White light cell (D$_2$O)

Dielectric mirror; elimination of 795nm

。www.utns.jp/~beam

Beam-Material Interactions

(UV –) VIS – IR

(<300 -) 533 - 2600nm

OPA
Preliminary Pulse Radiolysis; 795 nm

**Condition**

<table>
<thead>
<tr>
<th>Condition</th>
<th>H₂O&amp;1M H⁺</th>
<th>H₂O</th>
</tr>
</thead>
<tbody>
<tr>
<td>/ / mm</td>
<td>20 mm</td>
<td>5 mm</td>
</tr>
<tr>
<td>Charge</td>
<td>0.8-1.0nC</td>
<td>0.8-1.0nC</td>
</tr>
<tr>
<td>Beam size</td>
<td>4mm</td>
<td>4mm</td>
</tr>
<tr>
<td>Pulse width</td>
<td>7ps</td>
<td>3ps</td>
</tr>
<tr>
<td>Wavelength</td>
<td>795nm</td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>64</td>
<td>16</td>
</tr>
<tr>
<td>Time resol.</td>
<td>30ps</td>
<td>9ps</td>
</tr>
</tbody>
</table>

**Results**

- O.D. still low

In 1999

- Absorbance vs. Time (ps)
  - H₂O: Red line
  - 1M H⁺: Blue line

30ps

In 2002

- Absorbance vs. Time (ps)
  - 9ps

Intensity [a.u.]

Charge [a.u.]

www.utns.jp/~beam
**Pulse radiolysis using white light continuum**

- Time behaviors of $e_{aq}^-$ at 700nm

**Results**

<table>
<thead>
<tr>
<th>$l$ /mm</th>
<th>10</th>
<th>5</th>
<th>2</th>
<th>1</th>
</tr>
</thead>
<tbody>
<tr>
<td>O.D.</td>
<td>0.32</td>
<td>0.19</td>
<td>0.08</td>
<td>0.04</td>
</tr>
<tr>
<td>S/N</td>
<td>15</td>
<td>10</td>
<td>5</td>
<td>3</td>
</tr>
<tr>
<td>Dose</td>
<td>40Gy</td>
<td>47Gy</td>
<td>50Gy</td>
<td>50Gy</td>
</tr>
<tr>
<td>Time resol. /ps</td>
<td>12-13ps</td>
<td>6-7ps</td>
<td>4-5ps</td>
<td>&lt;4ps</td>
</tr>
</tbody>
</table>

**Time resolution:**

\[ \delta_{\text{total}} \approx \delta_{\text{diff}} + (\delta_E^2 + \delta_L^2 + \delta_{\text{sync}}^2)^{1/2} \]

Dominant factor: $\delta_{\text{diff}}$ due to refractive index $n=1.33$
**Comparison with conventional linac**

<table>
<thead>
<tr>
<th></th>
<th>Conventional linac</th>
<th>Photocathode</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Long-pulse mode</td>
<td>Single-pulse mode</td>
</tr>
<tr>
<td>Charge</td>
<td>8~20nC</td>
<td>0.5~0.6nC</td>
</tr>
<tr>
<td>Beam size</td>
<td>15x6mm</td>
<td>4mm</td>
</tr>
<tr>
<td>Dose/shot</td>
<td>30~50Gy</td>
<td>7~8Gy</td>
</tr>
<tr>
<td>Pulse width</td>
<td>10-100ns</td>
<td>10ps</td>
</tr>
</tbody>
</table>

**Dominant factors for time resolution**

1. <3ps : pulse width of EB
2. 100fs : pulse width of laser
3. <1ps : synch. jitter
4. 10ps /10mm : $\Delta t_{EB-L}$ in $H_2O$ → Thinner cell, but OD ↓ → **High-Brightness EB**

*Not so bad*
Femtosecond electron beam and femtosecond pump-probe experiment in Osaka University
A new concept of “equivalent velocity spectroscopy” for studies of ultrafast electron-induced reactions

Temporal distribution of 98 fs electron bunch measured by the streak camera at 0.17 nC

Charge: 0.17 nC

Transient absorption kinetics of hydrated electrons measured in water at wavelength of 800 nm

Intensity [a.u.]

Time [ps]

Optical density

Time [ps]
LEAF Facility Layout

LEAF

Accelerator Vault

Target A

(Area for future targets)

Probe Beam

798 nm

UV Beam

266 nm

Detectors

Electron Gun

Magnet Supplies

Control Console

Experimental Stations

Control Room

Laser Room

Klystron

RF System

Mechanical Chase

Anteroom

Stairway

Van de Graaff Room

(Hood)

13.7 m

20.7 m

2) Ti:Sapphire oscillator produces ~50 fs pulses, ~7 nJ energy, 798 nm, at 81.60 MHz.

3) Pulse stretcher stretches oscillator pulse to > 200 ps, then injects the pulse into the Ti:Sapphire regenerative amplifier.

4) Simultaneously, the doubled, Q-switched Nd-YAG laser pumps the Ti:Sapphire regen.

5) Stretched ~200 ps pulse is amplified to ~12 mJ level. Half is compressed to 1-3 ps for THG.

6) 1-3 ps pulse is frequency tripled to 266 nm (≤ 0.4 mJ) for excitation of Mg photocathode.

7) Half of regen output compressed to ~100 fs for use as probe or TOPAS OPA pump (8)
Pulse-Probe Experiment

- **Probe Beam**
  - Variable $\lambda$
  - 240 - 1700 nm

- **UV Beam**
  - 266 nm

- **Electron Beam**
  - Electron Gun

- **Detectors**

- **Faraday Cup**

- **Sample**

- **Delay**

- **Time, ns**

- **Pulse-Probe Experiment**
  - Water, 800 nm
  - 1 cm path
  - 9 ps FWHM

- **Electron Gun**

- **Pulse-probe controller**
  - Absorbance
  - Time, ps
  - Water, 800 nm
  - 5 mm cuvette
LEAF Pulse-probe transient absorption spectroscopy

Time resolution $\geq 7$ ps
A factor of pulse width and sample depth.

Optical Parametric Amplifier (240 - 2600 nm)
New diodes extend range from 1000 to 1700 nm
Color separation needs work, far-field mode varies

Better Signal/Noise than before
Improved LabVIEW acquisition software
Interleaved collection, measurement selection criteria (dose, laser intensity, modulator and laser timing), using consistent cathode recovery time
More proficient laser and electron beam alignment

Igor-based analysis software

Flow system volumes reduced ($\sim 4$ ml or $\geq 15$ ml)
ELYSE, Picosecond Pulse Radiolysis
ELYSE, Orsay

**Photoinjector Accelerator**

- Pulse length \( \leq 7 \) ps
- Charge \( \geq 1 \) nC
- Energy 4 to 9 MeV
- Repetition Rate \( \leq 50 \) Hz
- Energie Dispersion \( \leq 2,5 \) %
- Spot Diameter \( \leq 2 \) à 20 mm
- Pulse-Probe
New pulseradiolysis system

- Easy setup → Easy to experiment
- Quadropole for beam de-sizing → high time resolution
- Stabilizing white light → noise decreasing

[improvement]
Stability of Probe light intensity

Drum cell + Achromatic lens

Fluctuation 2.3%

Experimental results (right water)

O_D(720nm)final

Fig. 12  O.D.

previous (26[ps]): Current 8[ps]
<table>
<thead>
<tr>
<th>Institution</th>
<th>Beam Energy</th>
<th>Beam Current</th>
<th>Beam Width</th>
<th>Beam Size</th>
<th>Target Path Length</th>
<th>Synchronization</th>
<th>Laser Pulse Width</th>
<th>Total Time Resolution</th>
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</thead>
<tbody>
<tr>
<td>U. Tokyo</td>
<td>4+18=22MeV</td>
<td>2nC</td>
<td>1ps</td>
<td>3mm</td>
<td>1mm</td>
<td>&lt;1ps (rms)</td>
<td>100fs (532nm-2600nm) OPA (400-1100nm) white light made by Ti:Sa</td>
<td>3ps (white light)</td>
</tr>
<tr>
<td>LEAF, BNL, USA</td>
<td>9MeV</td>
<td>2-8nC</td>
<td>≥ 7 ps</td>
<td></td>
<td>10mm (right water)</td>
<td>Pico-sec.</td>
<td>100fs (240-2600nm) OPA</td>
<td>&gt;7ps (pulse-probe)</td>
</tr>
<tr>
<td>ELYSE, France</td>
<td>4 to 9 MeV</td>
<td>≥ 1 nC</td>
<td>≤ 7 ps</td>
<td>2-20mm</td>
<td></td>
<td></td>
<td></td>
<td>~7ps?</td>
</tr>
<tr>
<td>Waseda Univ.</td>
<td>4MeV</td>
<td>0.4-0.6nC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>8ps</td>
</tr>
<tr>
<td>Osaka Univ.</td>
<td>38MeV</td>
<td>&gt;0.2nC</td>
<td>&lt;1ps</td>
<td></td>
<td></td>
<td></td>
<td>100fs</td>
<td>~5ps</td>
</tr>
</tbody>
</table>
Summary

Photocathode RF gun with fs laser(Tt:Sa) is suitable combination for the Application of Radiation Chemistry.

In order to measure the phenomena at sub-pico or picosecond region, we need:

- high brightness beam with short pulse(<1ps)
- Thin target(~mm)
- Stable system
  - Timing (within 1ps)
  - Position
  - Beam Intensity (both laser and electron beam)
Special thanks to
LEAF, BNL
ELYSE, Orsay
Univ. Osaka
Waseda Univ.
Univ. Tokyo
Physicochemical stage

- Factor of the thermalization distance
  \[ \lambda_m = (n \sigma)^{-1} \]
  \[ \lambda = [-\lambda_m \ln (P \lambda_m)]/\alpha \]

- Factor of the cross section of geminate ion recombination
  \[ \sigma_{rec} \sim 3 \times 10^{-20} E^{-1.86} \times \beta \]

- Branching ratios in physicochemical stage stems from ionization and excitation

<table>
<thead>
<tr>
<th>Decay of the directly excited water molecules</th>
<th>Probability</th>
<th>Probability</th>
<th>Reaction No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂O*(A¹B₁) ( \rightarrow ) H₂O</td>
<td>P1</td>
<td></td>
<td>(1)</td>
</tr>
<tr>
<td></td>
<td>P2</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>P6</td>
<td></td>
<td>(4)</td>
</tr>
<tr>
<td></td>
<td>(1-P6)P1</td>
<td></td>
<td>(1)</td>
</tr>
<tr>
<td></td>
<td>(1-P6)P2</td>
<td></td>
<td>(1)</td>
</tr>
<tr>
<td>H₂O*(B¹A₁) ( \rightarrow ) H₂O⁺ + e⁻</td>
<td>P3</td>
<td></td>
<td>(2-1)</td>
</tr>
<tr>
<td></td>
<td>P4</td>
<td></td>
<td>(2-2)</td>
</tr>
<tr>
<td></td>
<td>P5</td>
<td></td>
<td>(2-3)</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Decay of the excited states results from the recombination</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂O* ( \rightarrow ) H₂O</td>
</tr>
<tr>
<td></td>
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<td></td>
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<td></td>
</tr>
</tbody>
</table>
Ultra-fast pump-and-probe pulse radiolysis study: radiation induced fast processes

Time behaviors of hydrated electrons in water: **Solvation time < time resolution < 10ps**

Time behaviors of solvated electrons in ethanol: **Observation of solvation process (e\(^{-}\)\(_{pre}\) \(\rightarrow\) e\(^{-}\)\(_{sol}\))**

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**Fs Ti:Sapp laser**

**Combination!!**

22MeV S-band electron linac

Laser photocathode

Compressor

Optical parametric amplifier (533-2600nm)

**PROBE!**

100fs, 533-2600nm

**PUMP!**

2-3ps, \(\sim\)2nC, 3mm\(\phi\)

---

**EtOH**

700nm

1100nm

1200nm

1300nm

1500nm

---

<table>
<thead>
<tr>
<th>Optical path</th>
<th>Absorbance</th>
<th>Time /ps</th>
</tr>
</thead>
<tbody>
<tr>
<td>10mm</td>
<td>0.35</td>
<td>~13ps</td>
</tr>
<tr>
<td>5mm</td>
<td>0.30</td>
<td>~6-7ps</td>
</tr>
<tr>
<td>2mm</td>
<td>0.25</td>
<td>4-5ps</td>
</tr>
<tr>
<td>1mm</td>
<td>0.20</td>
<td>&lt;4ps</td>
</tr>
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<table>
<thead>
<tr>
<th>Optical path</th>
<th>Absorbance</th>
<th>Time /ps</th>
</tr>
</thead>
<tbody>
<tr>
<td>700nm</td>
<td>0.12</td>
<td>0-20ps</td>
</tr>
<tr>
<td>1100nm</td>
<td>0.08</td>
<td>20-40ps</td>
</tr>
<tr>
<td>1200nm</td>
<td>0.06</td>
<td>40-60ps</td>
</tr>
<tr>
<td>1300nm</td>
<td>0.04</td>
<td>60-80ps</td>
</tr>
<tr>
<td>1500nm</td>
<td>0.02</td>
<td>80-100ps</td>
</tr>
</tbody>
</table>
**Data acquisition**

- Measurement of laser intensity and charge
  - B: Both beam and light  \( \rightarrow I_M(B) \text{ and } I_R(B) \)
  - L: Light only  \( \rightarrow I_M(L) \text{ and } I_R(L) \)
  - P: Beam only  \( \rightarrow I_M(P) \text{ and } I_R(P) \)
  - N: Neither beam nor light  \( \rightarrow I_M(N) \text{ and } I_R(N) \)
  - Charge  \( \rightarrow C \)

  \( (I_M: \text{Main light, } I_R: \text{Reference light}) \)

- Calculation of precise absorbance

  \[
  Absorbance = \log_{10} \frac{I_0}{I} = \frac{C_{\text{ave}}}{C} \cdot \log_{10} \left[ \frac{I_M(L) - I_M(N) \cdot I_R(B) - I_R(P)}{I_R(L) - I_R(N) \cdot I_M(B) - I_M(P)} \right]
  \]

  \( (C_{\text{ave}}: \text{Average of charges}) \)