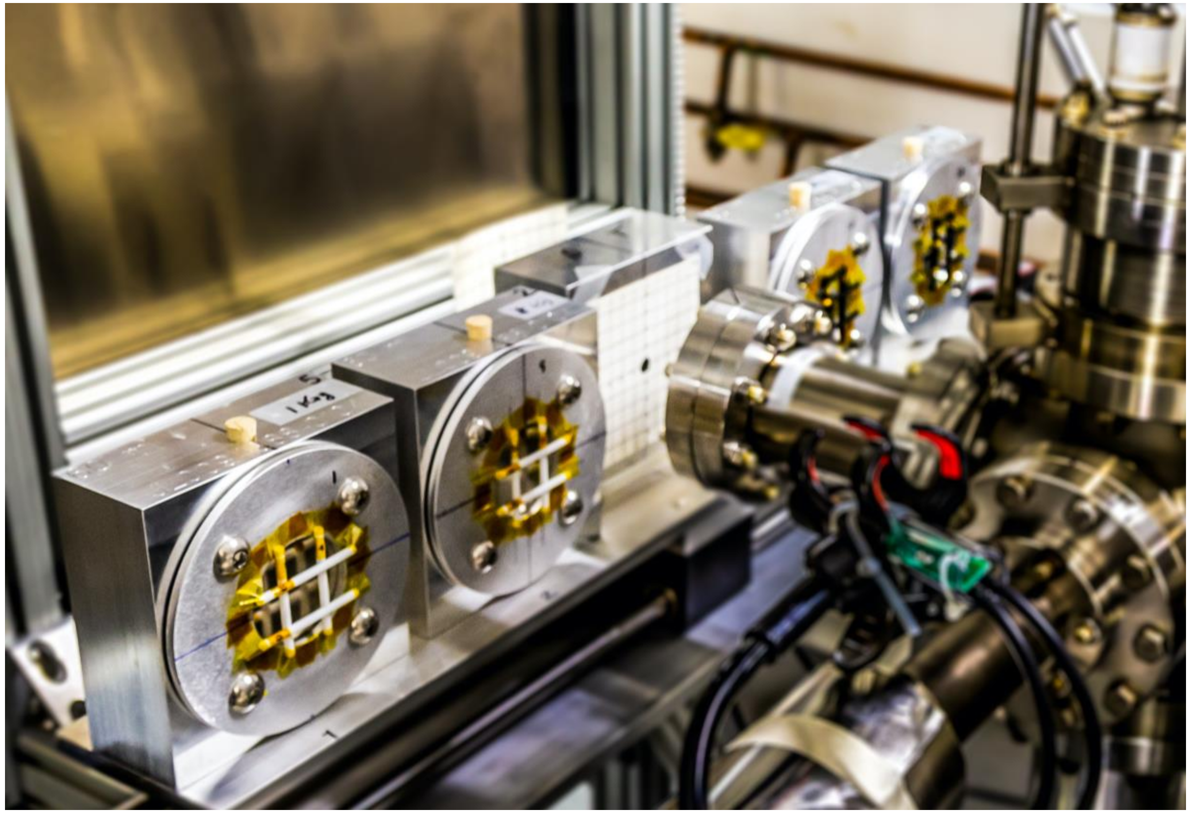
Electron Beam irradiation beamline at Jefferson Lab for 1,4-dioxane and PFAS remediation in Wastewater

NAPAC 2022



Xi Li, H. Baumgart, C. Bott, G. Ciovati, S. Gregory, M. McCaughan, F. Hannon, R. Pearce, M. Poelker, H. Vennekate, and S. Wang.

TUXD2

Presenter: Xi Li

Tuesday, August 9, 2022

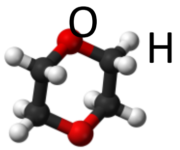
Outline

- Motivation
- Advantages and status of e-beam irradiation at wastewater plants
- E-beam irradiation beamline at Jefferson Lab: design and commissioning
- 1,4-dioxane and preliminary PFAS treatment results
- Challenges and possible solutions for wider adoption of e-beam technology
- Conclusions

Motivation

- Evaluate e-beam irradiation as a possible method to reduce or eliminate emerging contaminants in wastewater

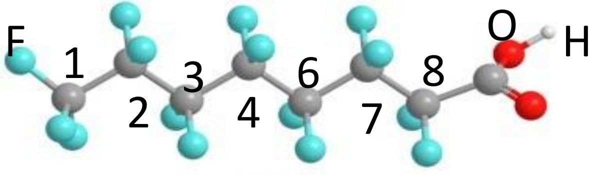
1,4-dioxane



- Widespread
- A likely human carcinogen

Perfluoroalkyl and polyfluoroalkyl substances (PFAS)

- A family of >5,000 synthetic substances



PFOA

Pollutant	Current method	Limitations
1,4-dioxane	Ozone	• Low effective degradation for lower concentration • ↑ **Bromate, toxic byproduct**
PFAS	Granular Activated Carbon (GAC)	• **Non-destructive** (separation) • Low degradation efficiency for low molecular weight PFAS

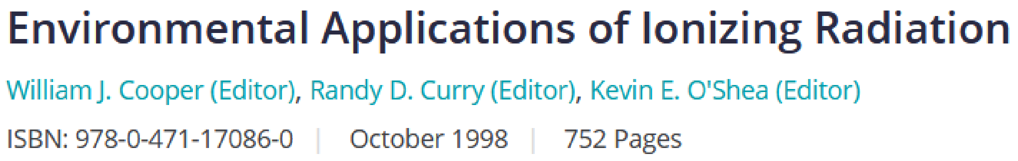
- These chemicals may soon be subjected to EPA regulation

Advantages of e-beam irradiation

Ionizing radiation

$$\underset{(1\text{ -}10\ \text{MeV})}{e^-} + H_2O \rightarrow \underset{\text{Oxidant}}{\cdot\,\mathbf{OH}(2.7)} + \underbrace{e^-{}_{aq}(2.5) + \cdot\,H(0.6)}_{\text{Reducers}} \ldots$$

- Production of both oxidizing and reducing species
- No need for chemical additives
- Proven effective in decomposing a wide range of organic chemicals with a relatively low dose (< ~2 kGy). Many such pioneering studies were done in the US in the 1980-90s



Environmental Applications of Ionizing Radiation

William J. Cooper (Editor), Randy D. Curry (Editor), Kevin E. O'Shea (Editor)

ISBN: 978-0-471-17086-0 | October 1998 | 752 Pages

Examples of e-beam irradiation at wastewater plants

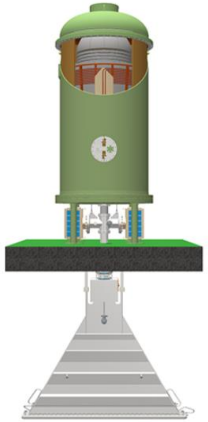
- Daegu dyeing treatment plant, South Korea, 10,000 m^3/day, 2006
- Guanhua knitting factory treatment plant, Guangdong, China, 30,000 m^3/day, 2020
- Medical waste treatment plant, Shiyan, China, 400 m^3/day, 2021

https://www.ans.org/news/article-3073/chinas-electron-beam-technology-for-treating-industrial-wastewater/

S. Wang, et al, Radiation Physics and Chemistry 196 (2022) 110136

Commercial accelerators: Transformer/DC type. Relatively compact, high efficiency but low beam power

ELV-type: 0.5 – 3 MeV, 20 – 400 kW



https://www.eb-tech.com/en/

South Korea

Dynamitron: 0.5 – 5 MeV, 30 – 200 kW

http://www.dasheng.com/en/dd.html

China

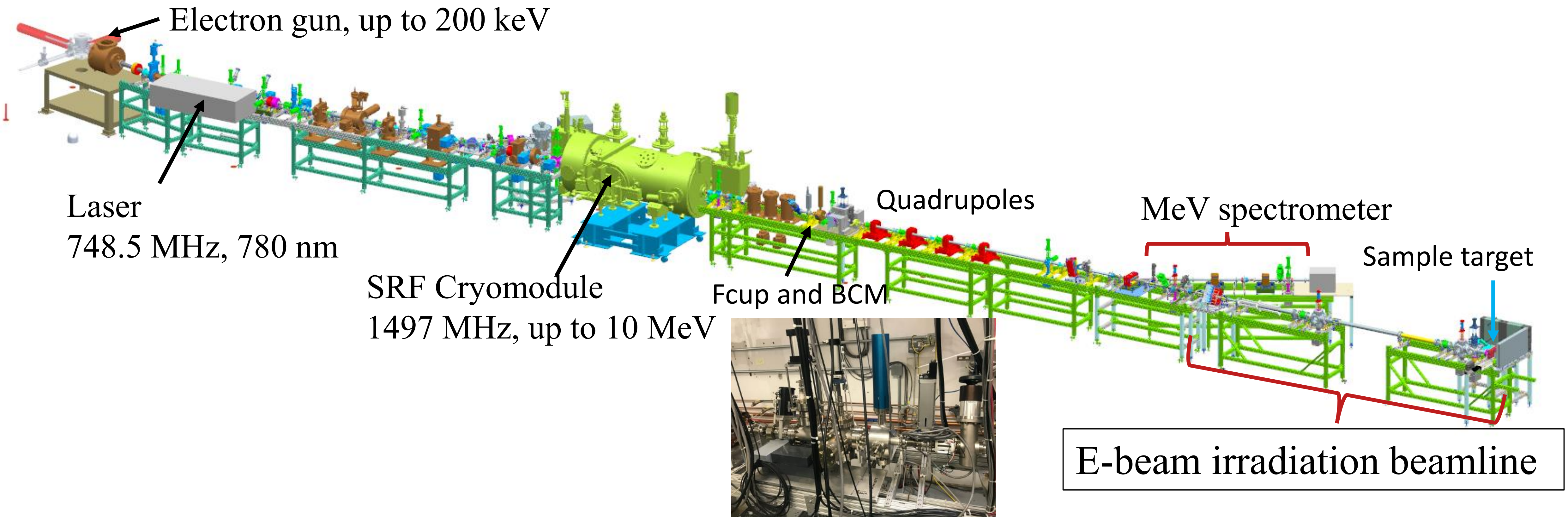
Jefferson Lab – HRSD collaboration

- Hampton Roads Sanitation District (HRSD)
  - Regional wastewater treatment utility company serving Southeast Virginia
  - Forward thinking, engaged in R&D on novel treatment techniques
  - Operates an R&D facility processing wastewater to drinking water standards, aiming at recharging the local aquifer (https://www.hrsd.com/swift)
  - Provided all the water samples and directed the analysis before and after irradiation

- Jefferson Lab:
  - Designed and built an irradiation beamline at a 10 MeV SRF accelerator (UITF) on JLab's campus
  - Became one of a handful of facilities where e-beam irradiation studies can be done in the US

The Upgraded Injector Test Facility (UITF) at Jlab

- Multi-purpose SRF continuous-wave (CW) accelerator
- Up to 10 MeV, 100 nA (limited by radiation shielding)



Electron gun, up to 200 keV

Laser
748.5 MHz, 780 nm

SRF Cryomodule
1497 MHz, up to 10 MeV

Fcup and BCM

Quadrupoles

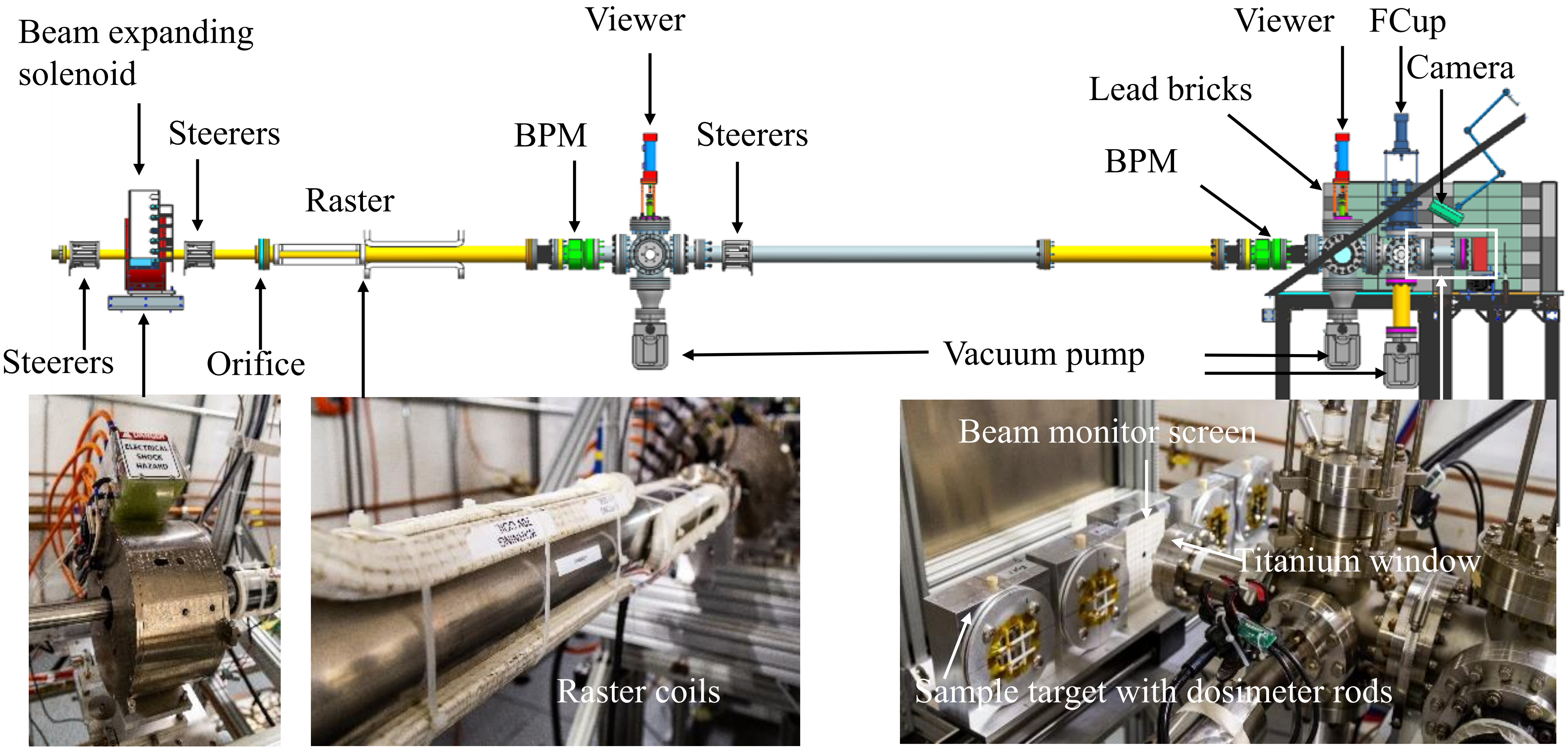
MeV spectrometer

Sample target

E-beam irradiation beamline

E-beam irradiation beamline

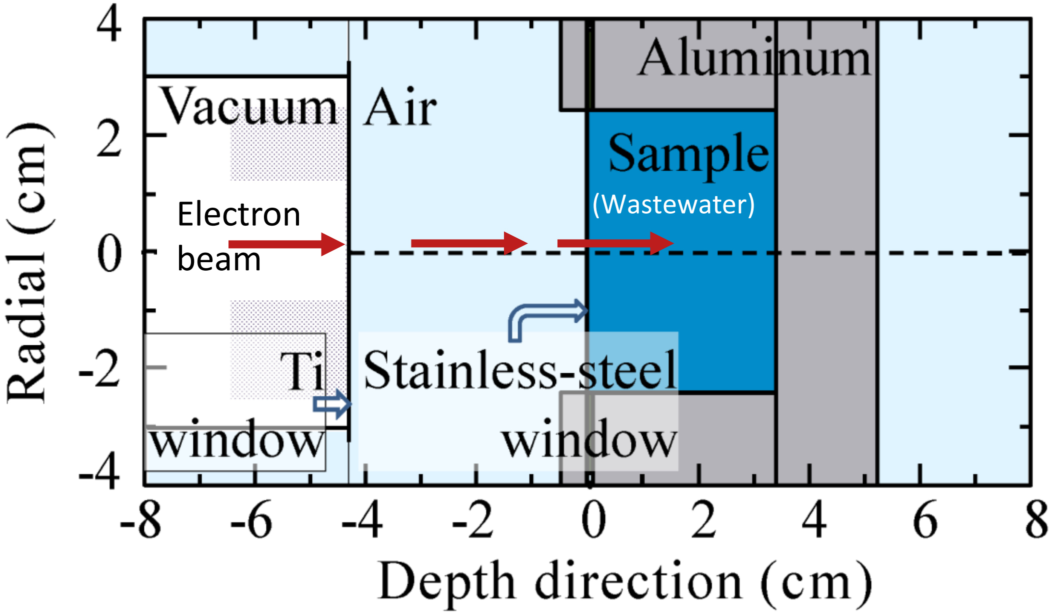
Xi Li *et al.*, Nuclear Inst. and Methods in Physics Research, A **1039** (2022) 167093



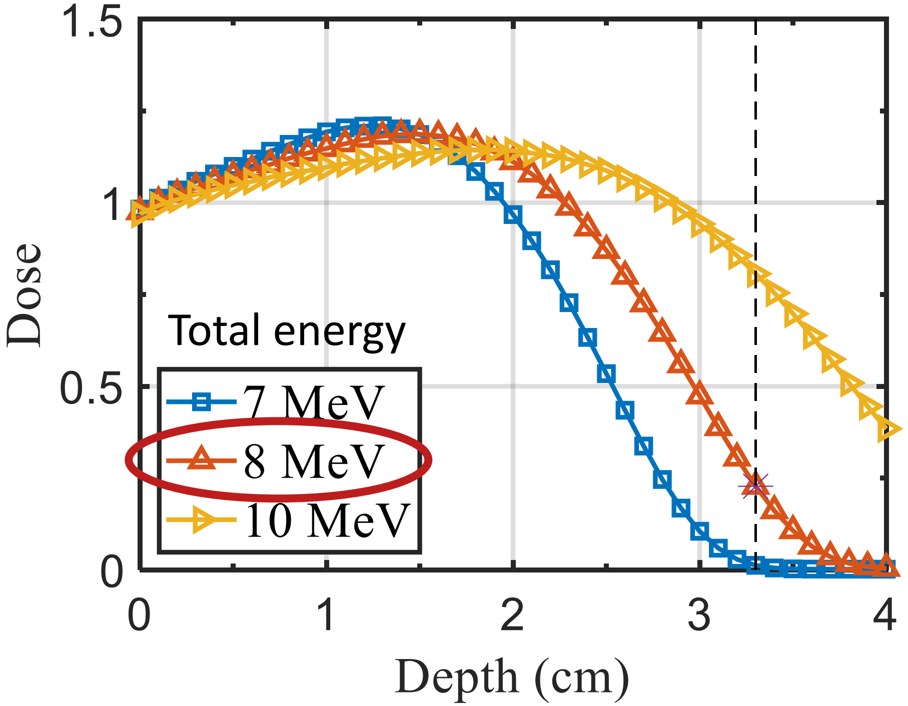
Beam parameter design

- Sample diameter = beam transverse diameter ≅ 50 mm
- Sample volume ≅ 60 mL -> beam energy = 8 MeV, a compromise between dose uniformity and reliable beam energy

Schematic layout in Monte-Carlo (FLUKA) simulation

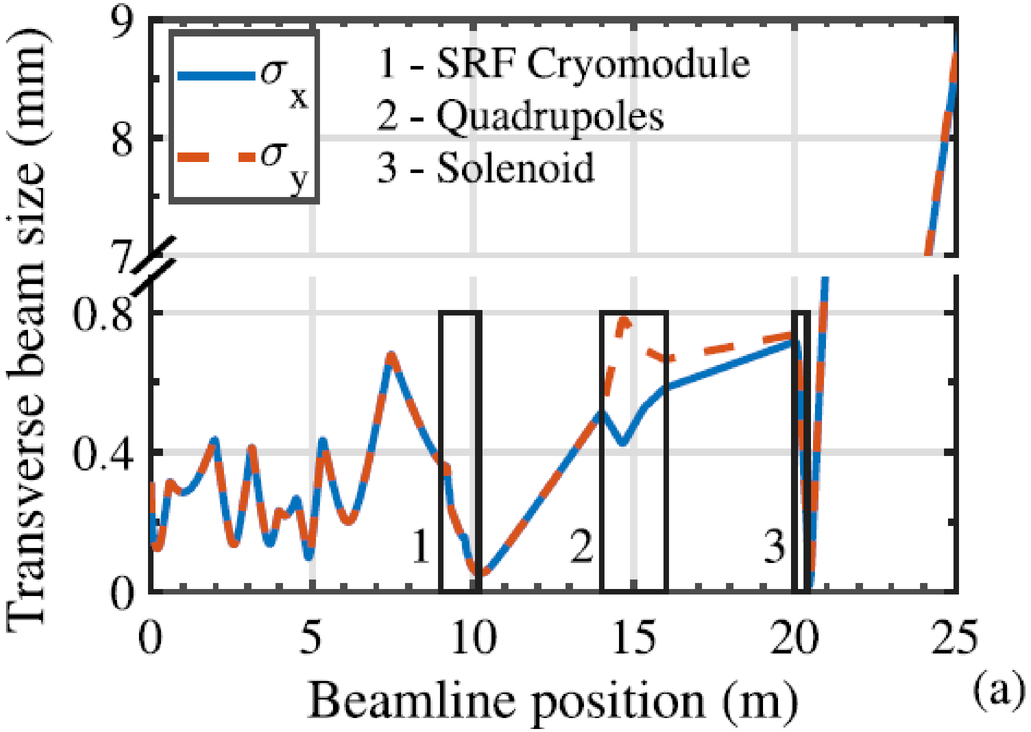


Window thickness: 0.127 mm
Ti: titanium



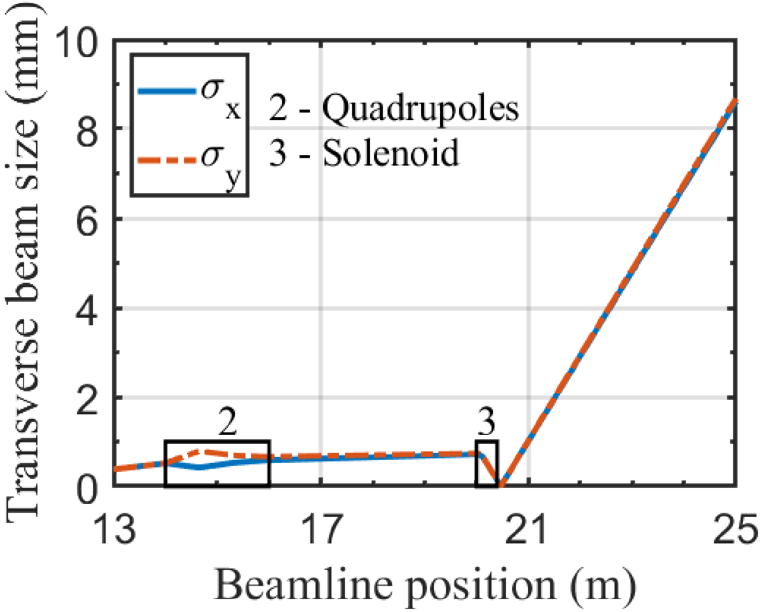
Beam envelope simulation by General Particle Tracer (GPT)

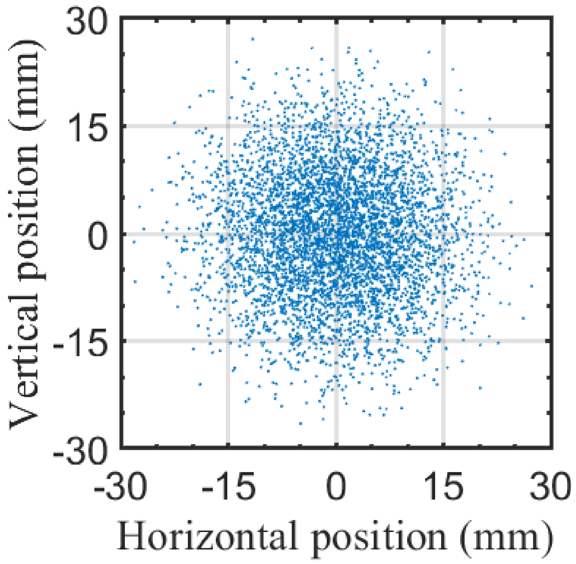
Parameters at target location	**Value**
Beam transverse size, σ	≅ 15 mm (90% electrons in the diameter of 50 mm)
Energy	8 MeV
Divergence	Simulation range < 10 mrad
Relative energy spread (σ_E/E)	Simulation range < 10^{-2}

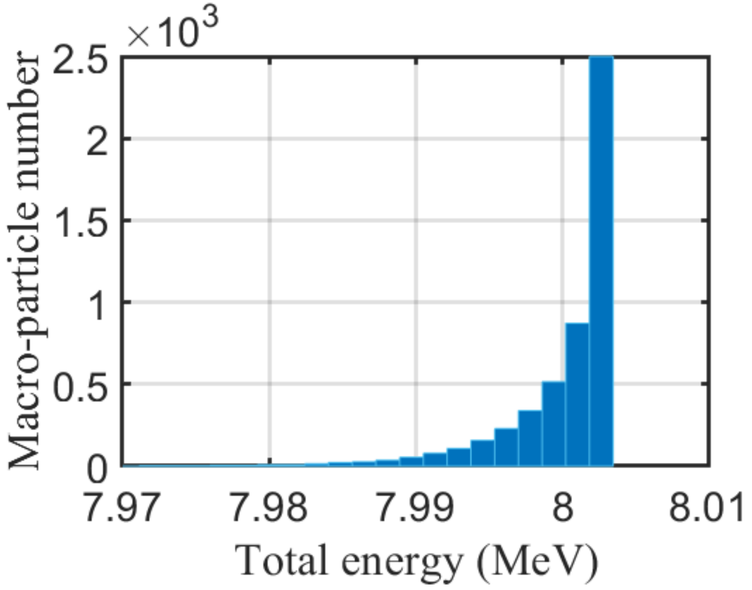


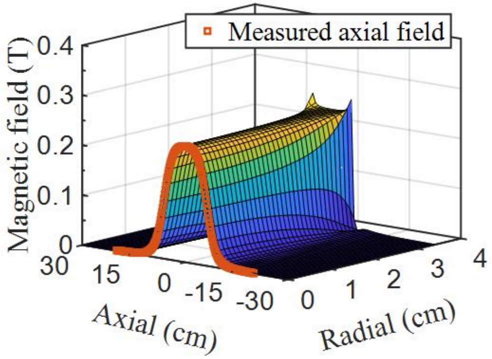
Beam transport along the beamline

- Use a solenoid with 0.28 T axial field to defocus the beam









σ: ~ 8.5 mm

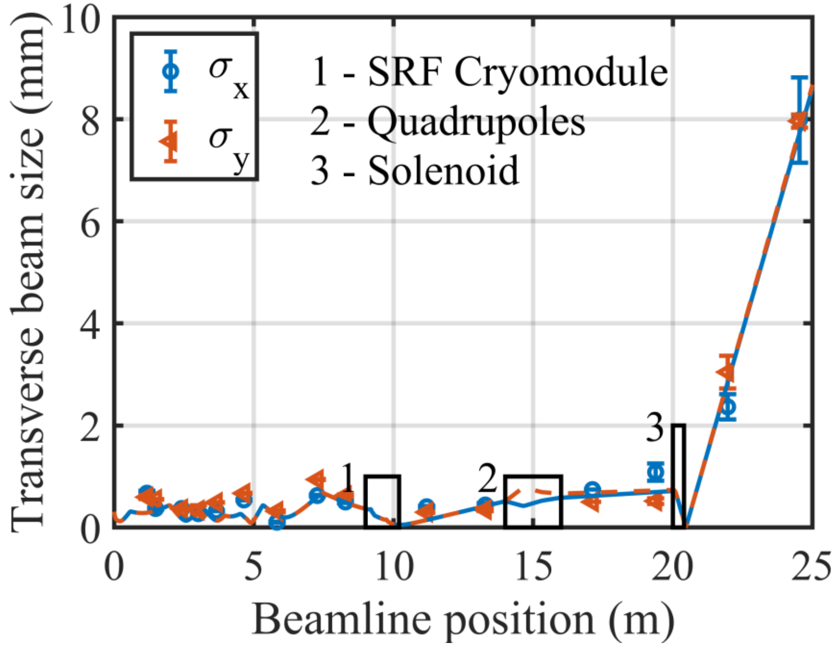
Relative energy spread: ~ 10^{-4}.

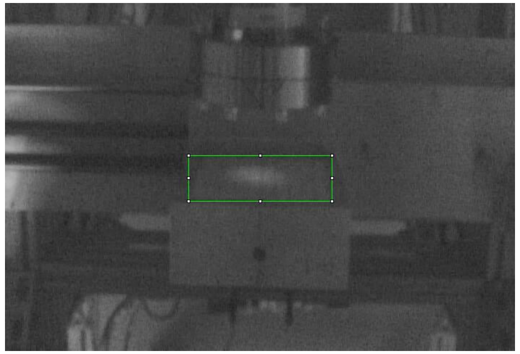
Space charge effect: beam doesn't have significant change

Add raster coils to further increase σ

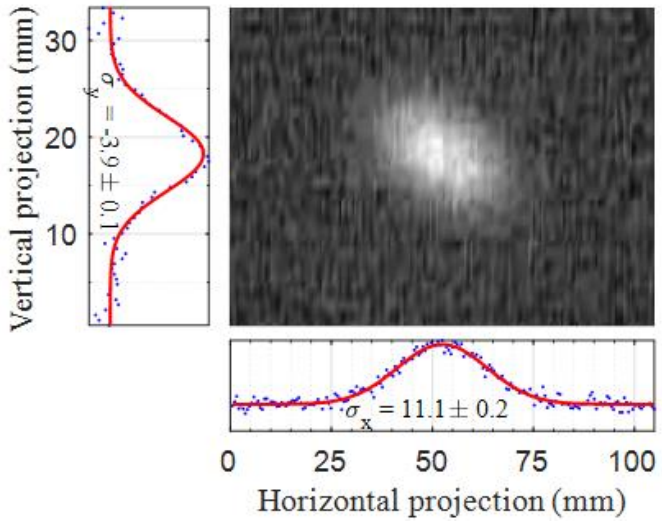
Beamline commissioning

- Beam size (1σ) is about 15 mm at the dummy target.

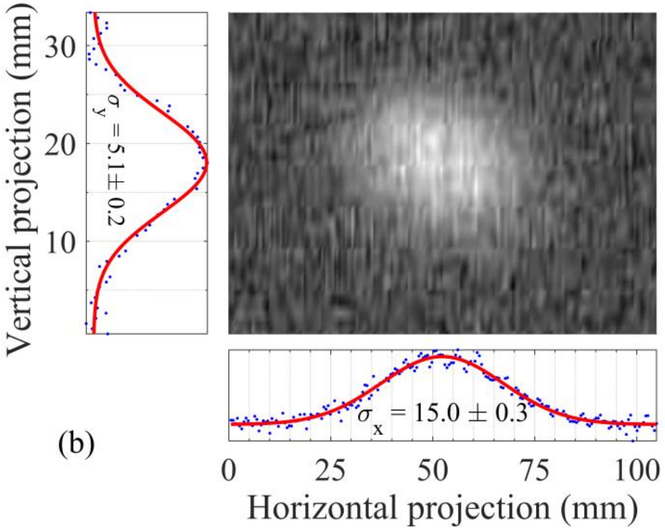


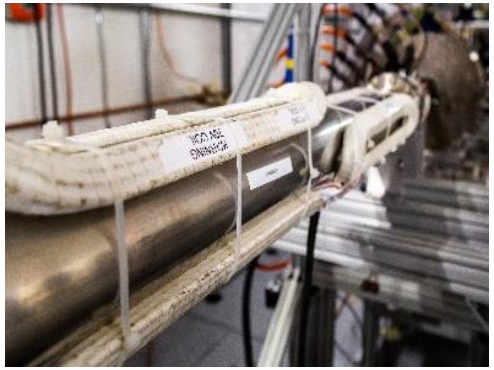

Beam on dummy target

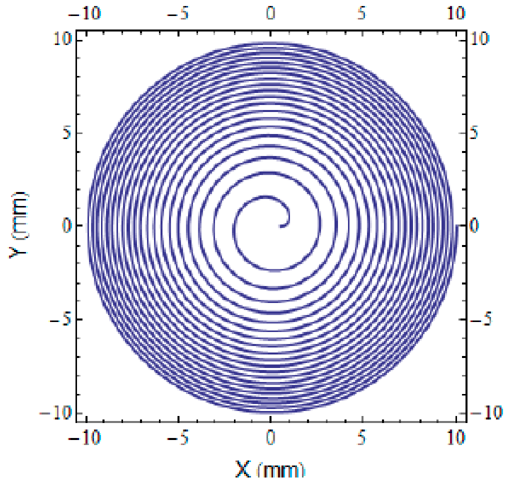
Raster off



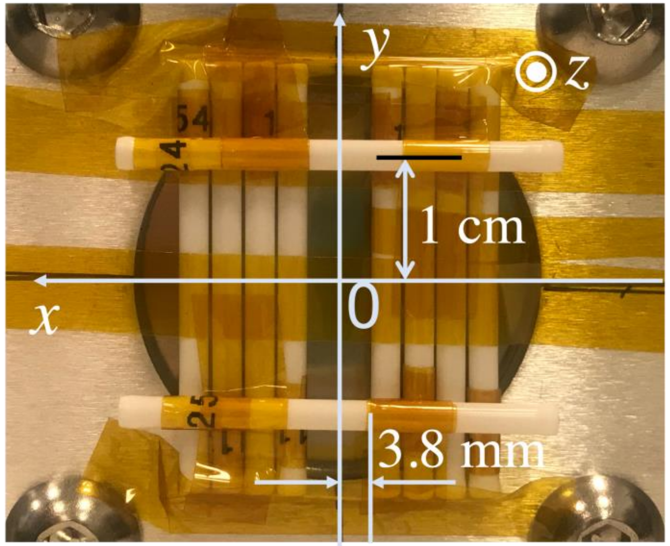
Raster on

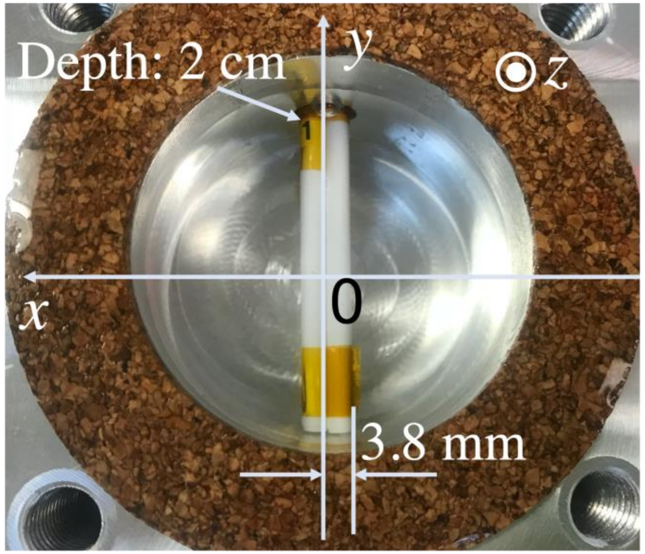


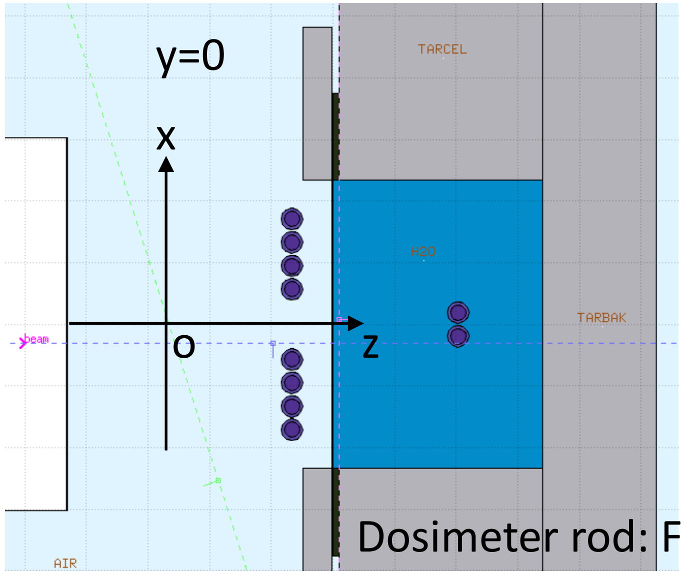




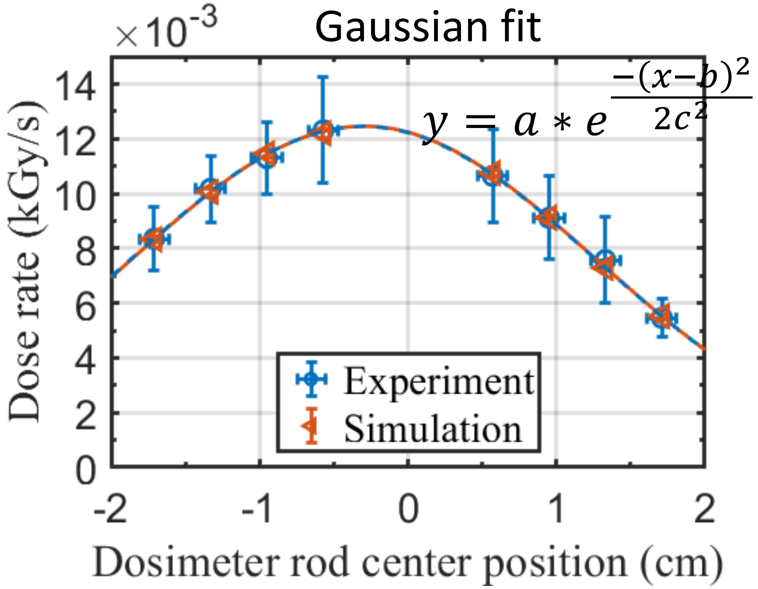
Dose distribution methodology







Dosimeter rod: FWT 70-40M

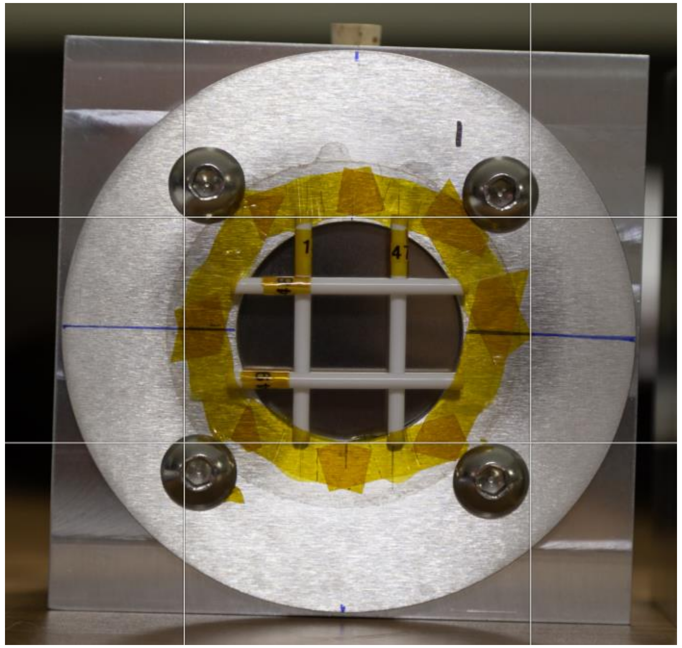


$$y = a * e^{\frac{-(x-b)^2}{2c^2}}$$

- Use experiments to calibrate the Monte-Carlo simulation
- Use simulation to calculate the dose distribution in the entire sample

Beam size, 1σ	**15.3 mm**
Beam center	(-3.0 , 1.7) mm
Total energy	8 MeV
Beam current	108 nA

Sample irradiation – dose distribution methodology



- 4 dosimeter rods mounted at the front of the target cell to monitor the dose distribution during sample irradiation

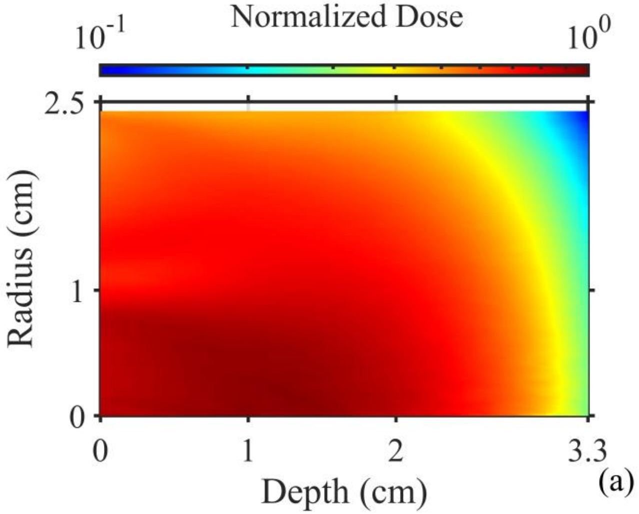
Delivered dose

Irradiation time

$$D = I_b D_t t$$

Beam current

Calibrated simulated dose rate per incident electron

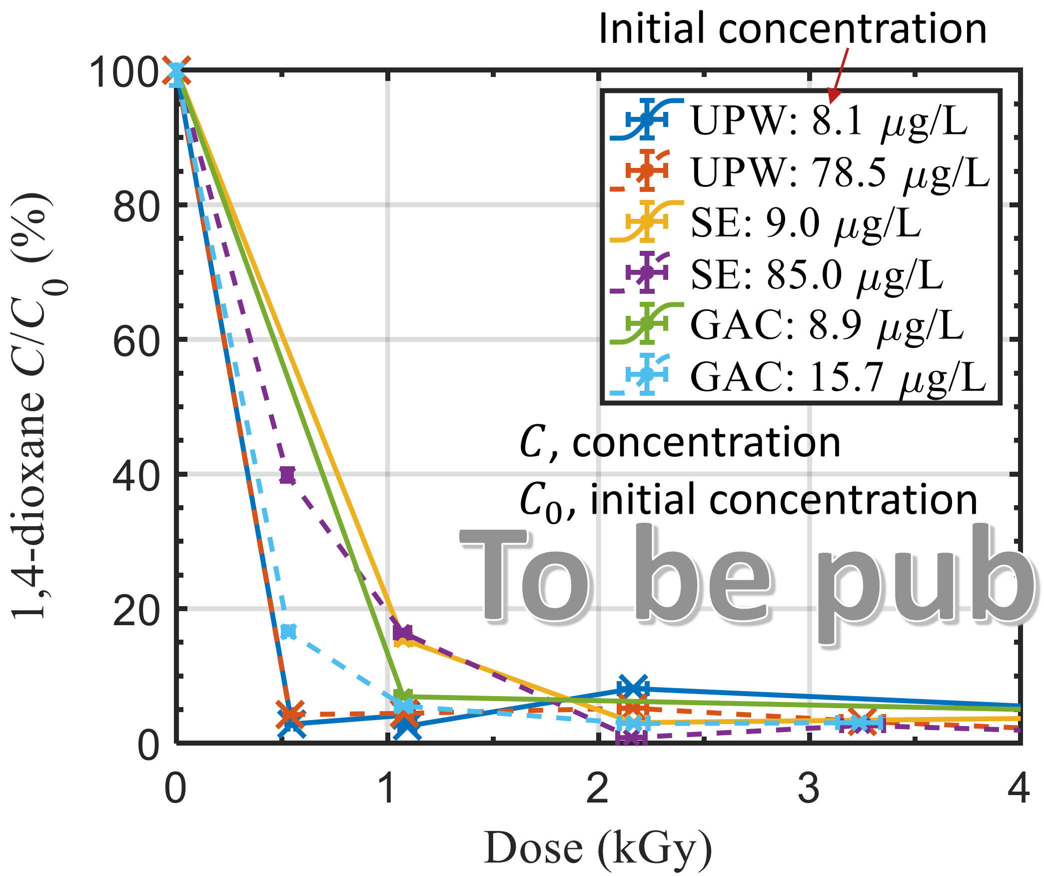


Dose distribution within the water volume, calculated with FLUKA

1,4-dioxane and PFAS treatment results

- More than 95% of 1,4-dioxane was removed for a dose < 2 kGy

- Conversion of long-chain PFAS (PFOS) to short-chain type (PFBA). Samples analyzed by Eurofins.

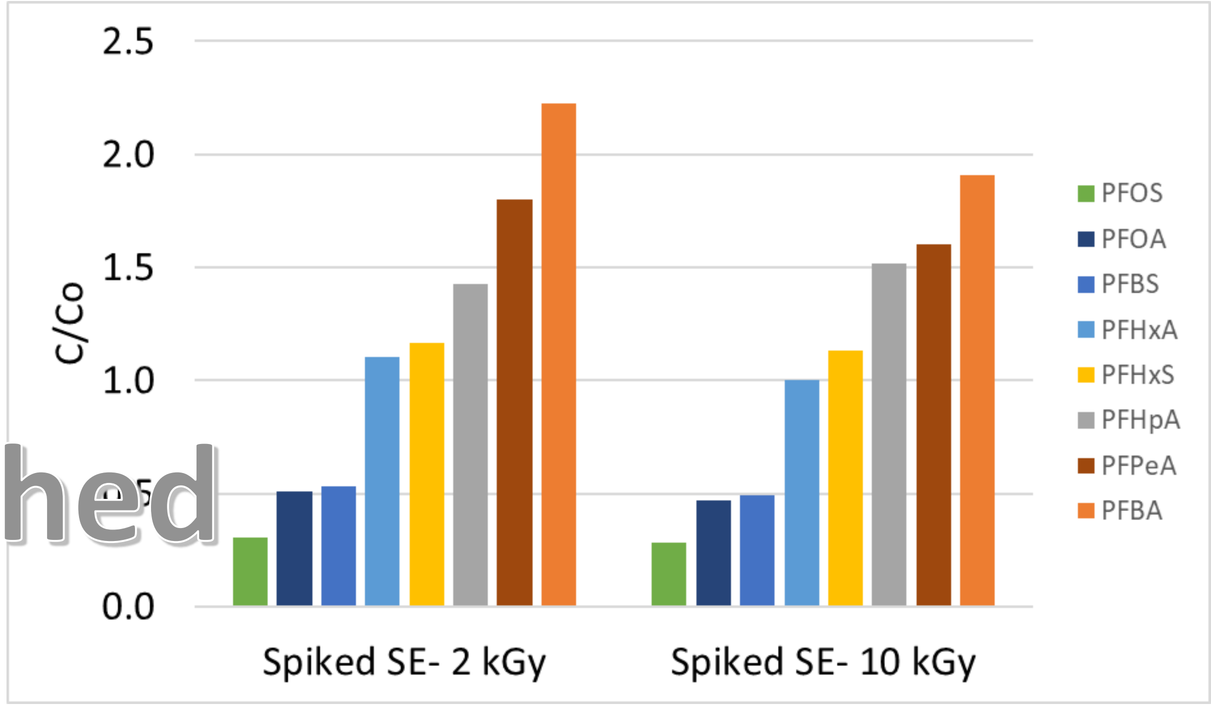


Initial concentration

C, concentration

C_0, initial concentration

UPW: ultra-pure water, SE: secondary effluent
GAC: granular activated carbon filtered SE
Analysis method: solid phase extraction



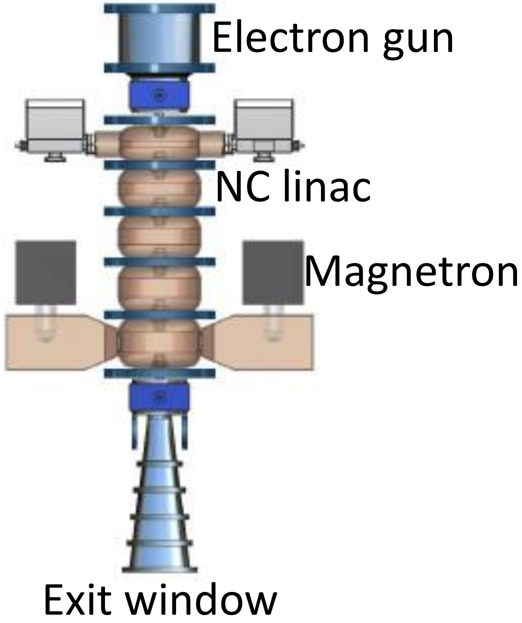
Spiked samples initial concentration
PFOS: ~ 500 ng/L, PFOA: 1 μg/L, PFHxA: ~ 500 ng/L

Challenges towards wider adoption of e-beam irradiation in wastewater treatment

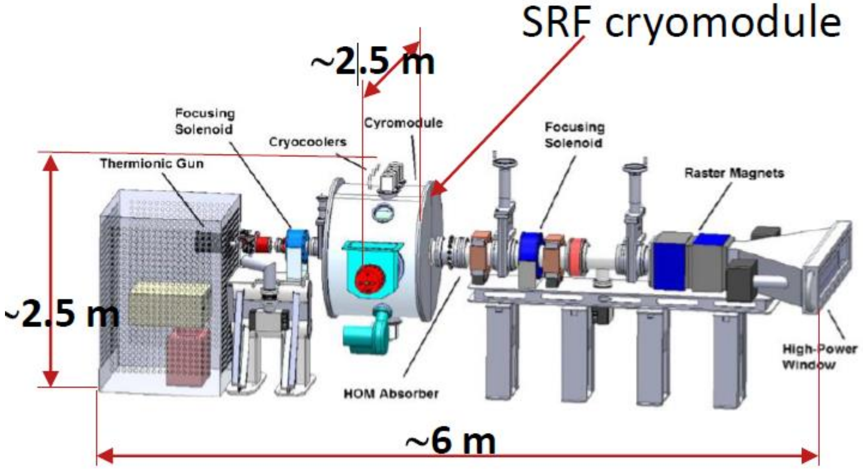
- $\text{Beam power} \propto \text{Mass flow} * \text{Dose}$. Accelerators with **higher beam power** and similar **high efficiency** than currently available need to be developed to achieve a **competitive treatment cost** in large-scale treatment plants. They should also be “**compact**” and have a **high reliability**.
- Only two vendors are producing the accelerators currently being used for wastewater treatment (none in the US)
- Education: e-beam irradiation is not a method typically mentioned in wastewater treatment engineering textbooks
- Conservative, well-established industry in developed countries, more open to innovation in developing countries (increasing fraction of wastewater being treated there)
- Funding to develop and improve accelerator technology for this application is fairly limited

SRF/NC-RF compact accelerator designs for wastewater treatment

1 MeV, **10 – 500 kW, 915 MHz**

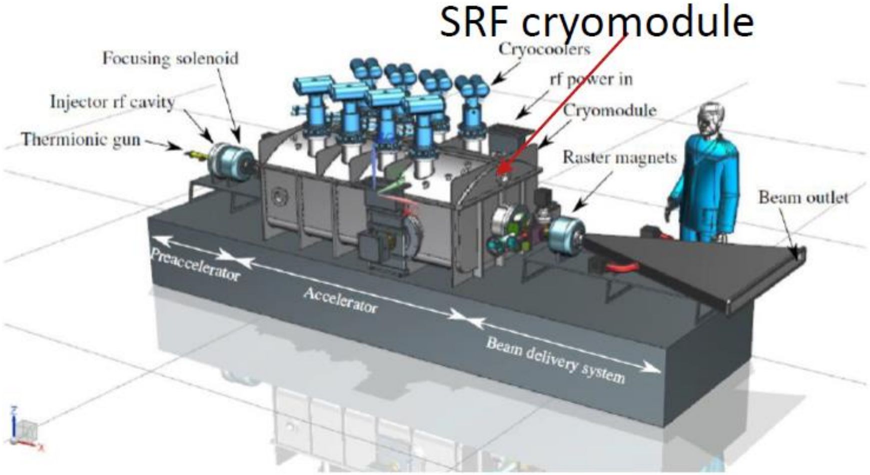


1 MeV, **1 MW, 750 MHz**



G. Ciovati *et al.*, *Phys. Rev. Accel. Beams* **21**, 091601 (2018)

10 MeV, **1 MW, 650 MHz**



R.C. Dhuley, *et al., Phys Rev. Accel. Beams* **25**, 041601 (2022)

Compact, High-Power SC Electron LiINACS for MW Industrial Applications,
C. Thangaraj (FNAL) **WEZE3**

Conclusions

- A collaboration between JLab and HRSD was initiated to study the effectiveness of e-beam irradiation towards treating emerging harmful contaminants in wastewater
- An e-beam irradiation beamline was successfully designed and commissioned at a 10 MeV SRF accelerator on JLab’s campus
- Successfully demonstrated >95% removal of 1,4-dioxane with < 2 kGy
- More complex chemistry with PFAS irradiation, more studies are needed
- Several challenges are preventing a wider use of e-beam irradiation as part of the treatment-chain in wastewater plants
- Advances in accelerator technology may hold the solution to some of those challenges.

Acknowledgements

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- Raster: B. Gunning, W. Lu
- Electric gun: G. Palacios-Serrano, C. Hernandez-Garcia
- Solenoid field maps: J. Grames
- Peppo solenoid: N. Falls, M. Beck
- Mechanical: S. Gregory
- Dosimeter rod and reader: D. Hamlete, K. Welch
- FLUKA simulation: K. Welch, FLUKA cern experts and the user forum
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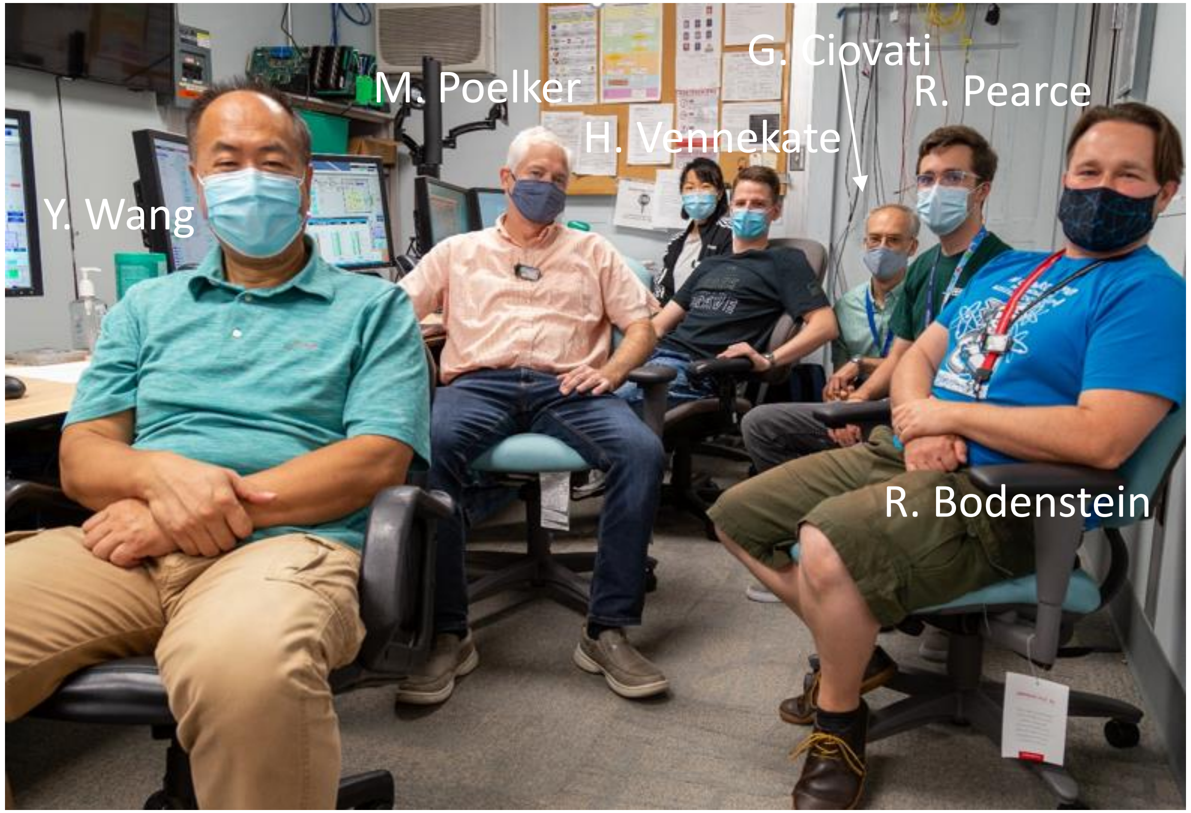
- SRF cavity: H. Wang



Thank you and Questions?