

Isotope Harvesting Research at FRIB

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MICHIGAN STATE UNIVERSITY

U.S. DEPARTMENT OF ENERGY | Office of Science

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Overview

- Introduction
- Isotope Harvesting Experiments
  - Aqueous harvesting of ^{62}Zn parallel to gas phase ^{76,77}Kr collection
  - Aqueous phase harvesting of ^{47}Ca & ^{48}V
  - Solid phase harvesting
- Radiolysis phenomena in a flowing-water target
  - Experimental results
  - Simulation model & upscaling towards FRIB conditions
- Source sample preparation for BMIS
  - Stable (^{30}Si, ^{50}Cr, ^{58}Fe) & long-lived (^{7}Be, ^{26}Al, ^{32}Si) sources

Rare Isotope Production at NSCL & FRIB and Isotope Harvesting

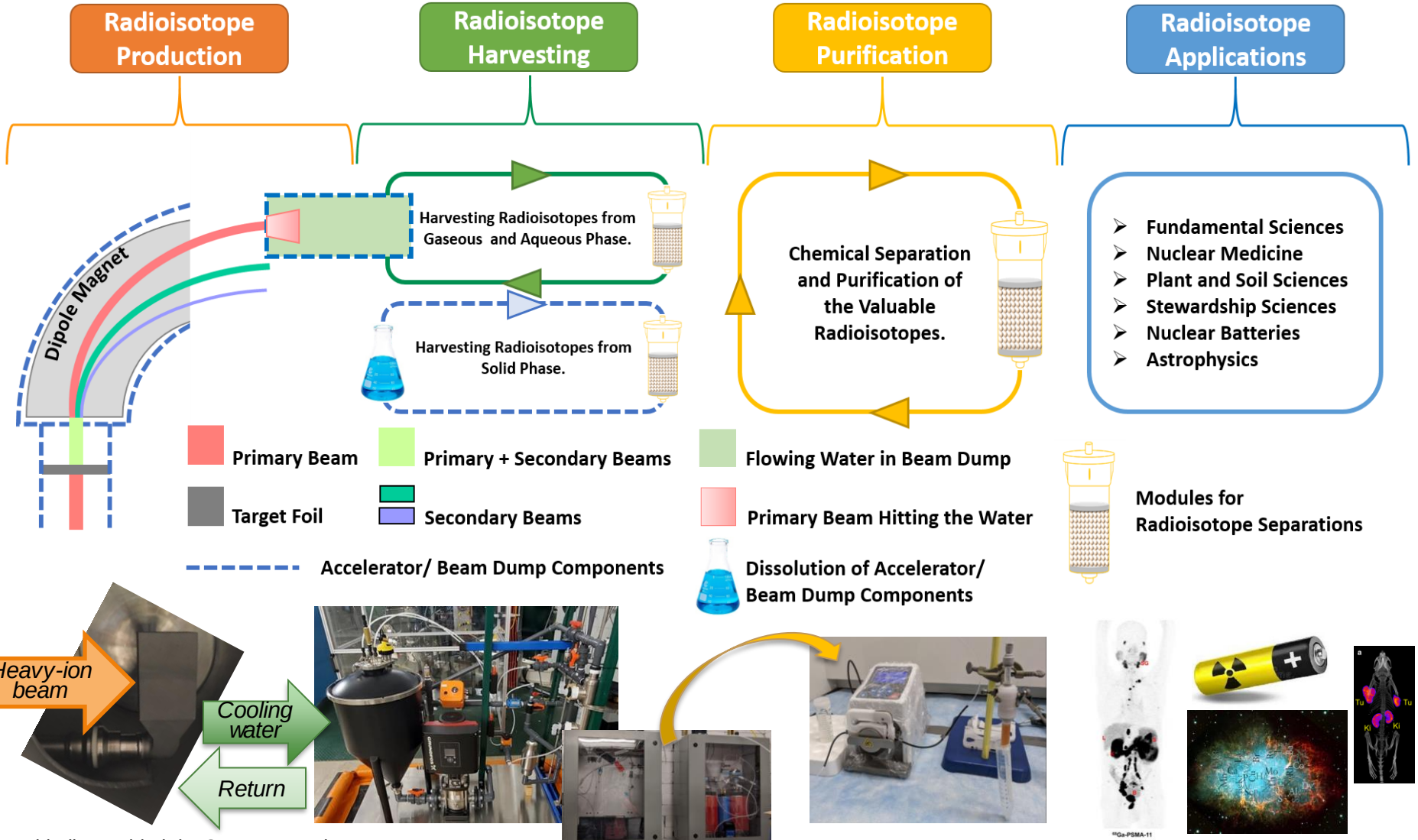
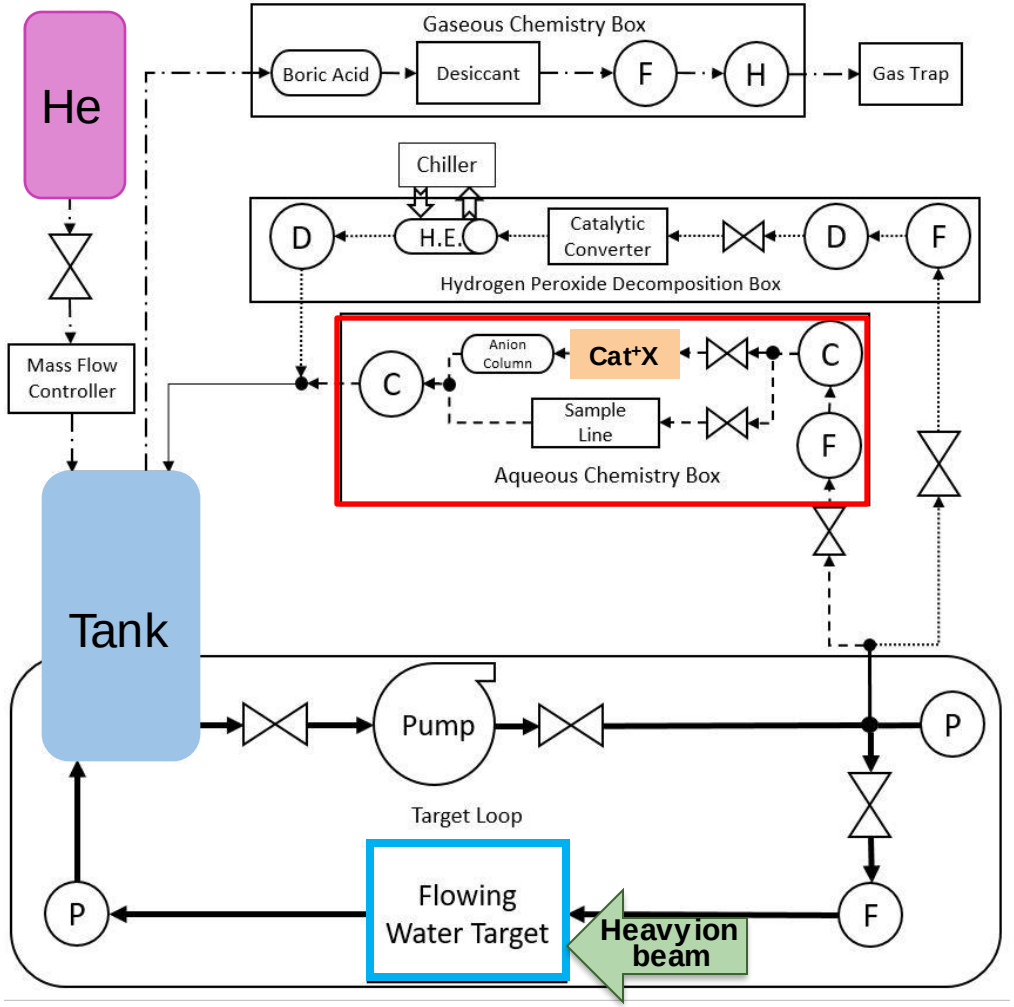


Image kindly provided by S. Essenmacher

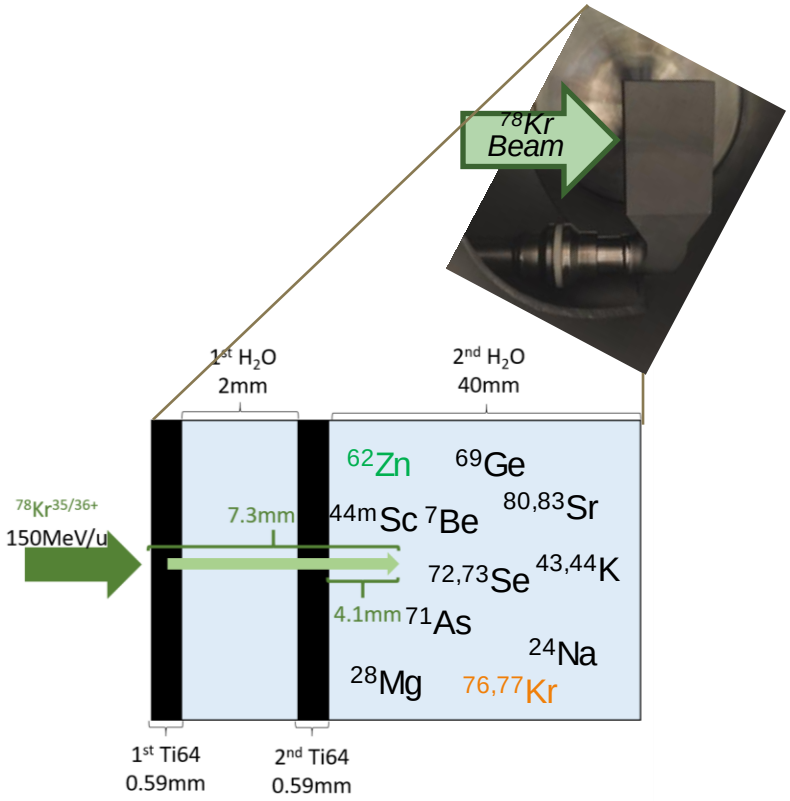
Experimental Setup for Isotope Harvesting at NSCL



References: Abel *et al.* (2020), Domnanich *et al.* (2020)

Two ^{78}Kr-beam experiments

- t_{irr}: 5.5 – 7.8 h
- Beam power: 2.0 – 25.1 W



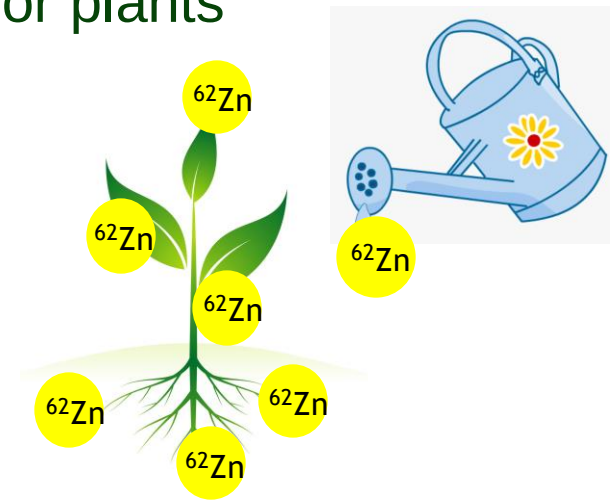
Why is ^{62}Zn interesting?

- Nuclear medicine: ^{62}Zn/^{62}Cu PET generator

$$^{62}\text{Zn} \xrightarrow{\beta^+/\text{EC}} {}^{62}\text{Cu} \xrightarrow{\beta^+} {}^{62}\text{Ni (stable)}$$

	^{62}Zn	^{62}Cu
Half-life	9.2 h	9.7 min
Decay mode	7% β^+, 97% EC	98% β^+, 2% EC
Main γ-rays [keV]	40.8 (25.5%), 507.6 (14.8%), 549.4 (15.3%), 596.6 (26.0%)	1173 (0.3%)

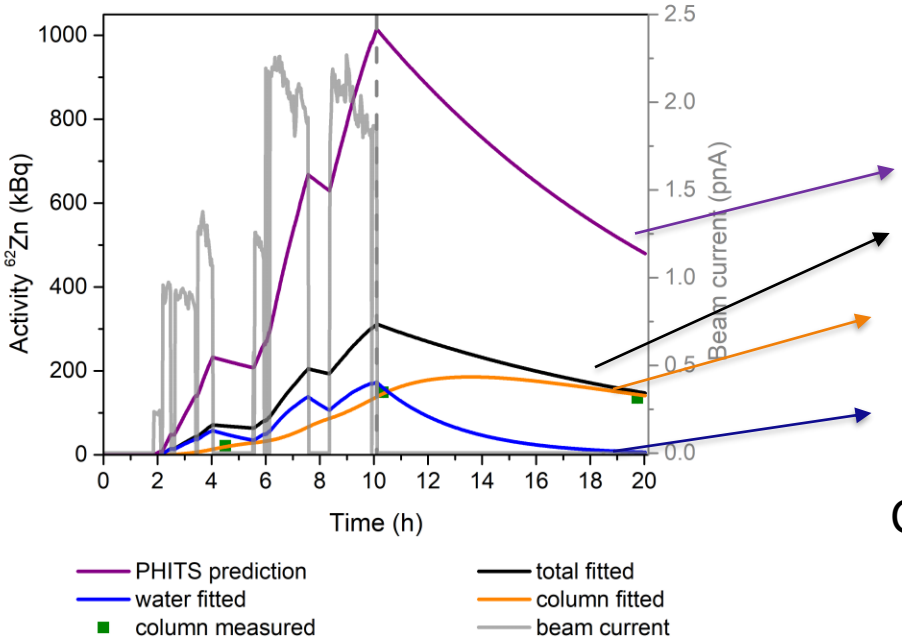
- Zn: essential nutrient for plants
  - Plant uptake studies



References: Almas *et al.* (2001), Ng *et al.* (2014)

Collection of ^{62}Zn

Mass transport in isotope harvesting system



	Production rate [^{62}Zn/incoming ^{78}Kr]	A [kBq]
PHITS	3.07*10^{-4}	1020
Experiment	9.08(30)*10^{-5}	312

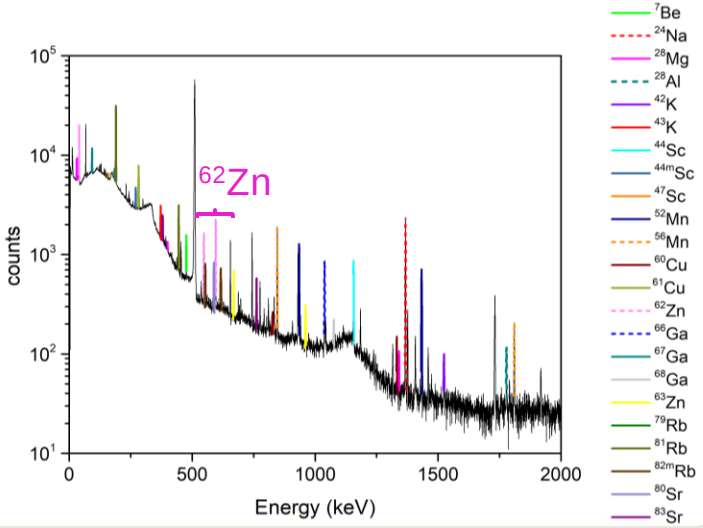
$$\frac{dN_{column}}{dt} = k_{ads}N_{water} - \lambda N_{column}$$

$$\frac{dN_{water}}{dt} = P * I(t) - k_{ads}N_{water} - \lambda N_{water}$$

Collection efficiency: ~97%

Chem. purification of ^{62}Zn

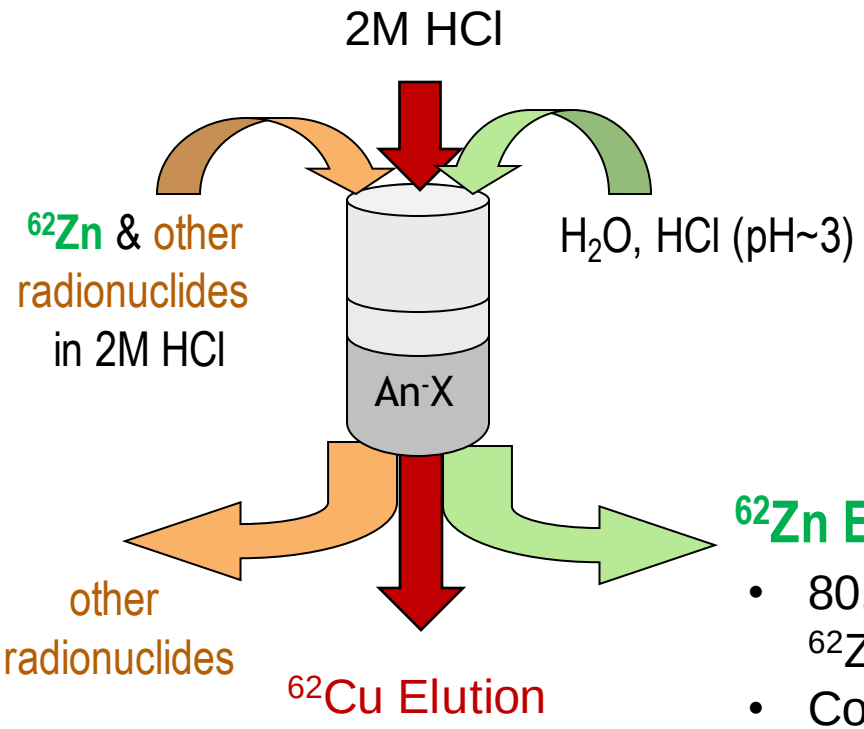
- Elution from Cat^{+}X column
- Co-elution of other by-produced radionuclides
  - ^{7}Be, ^{24}Na, ^{28}Mg, $^{44g/m,47}$Sc, ^{52,56}Mn, ^{61}Cu, ^{63}Zn, ^{66,67}Ga, ^{81,82m}Rb, ^{81,83}Sr



Reference: Domnanich *et al.* (2020)

Separation of ^{62}Zn & ^{62}Zn/^{62}Cu Generator

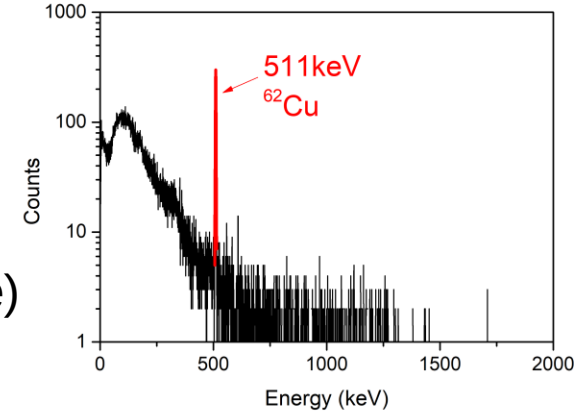
Separation on An^{-}X Column



Pure ^{62}Cu eluate

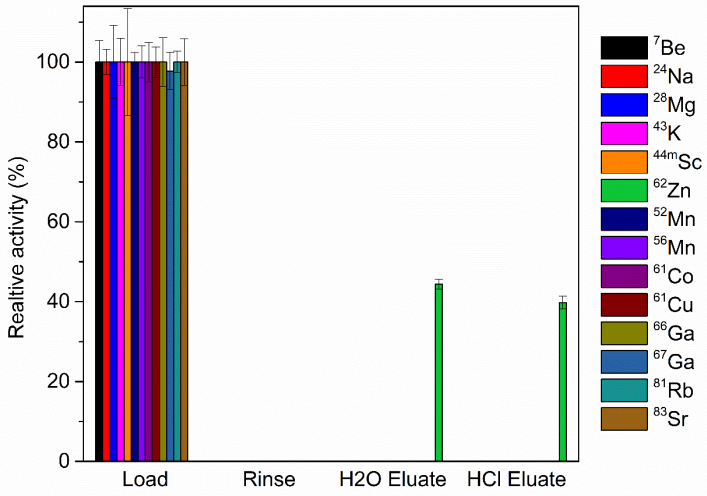
- Fast generation of ^{62}Cu

^{62}Zn *9.2h* → ^{62}Cu *9.7min* → ^{62}Ni (stable)



^{62}Zn Elution

- 80.5% overall ^{62}Zn yield
- Collection from harvesting system & isolation



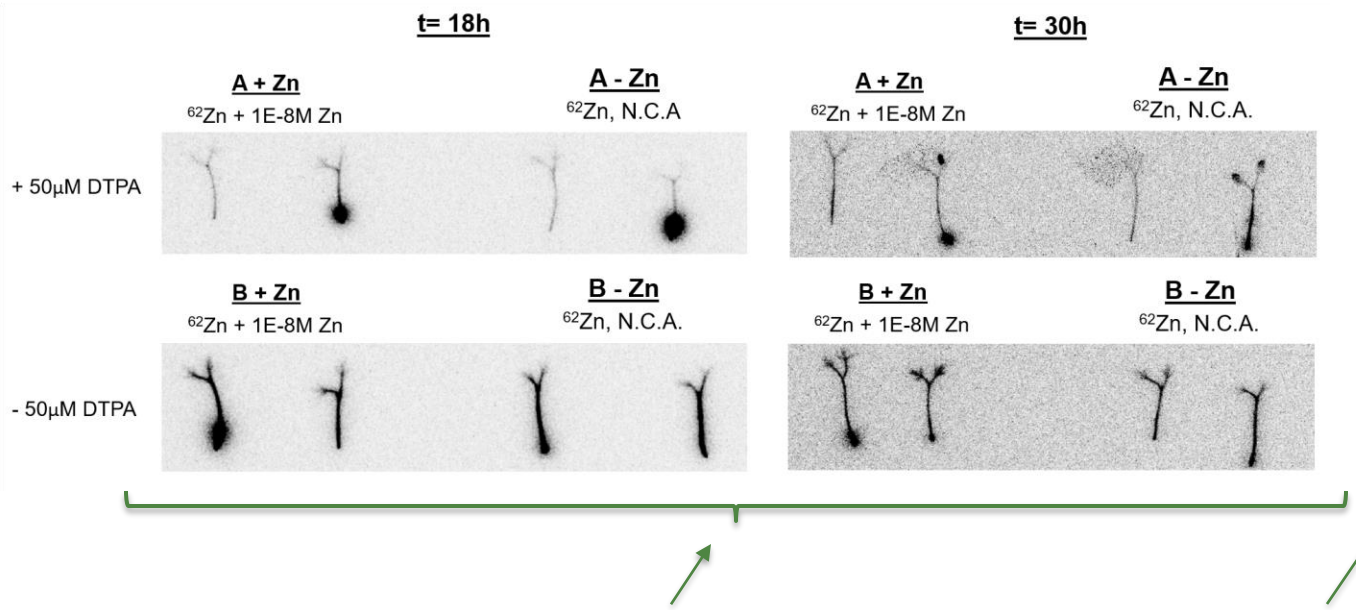
Reference: Domnanich *et al.* (2020)

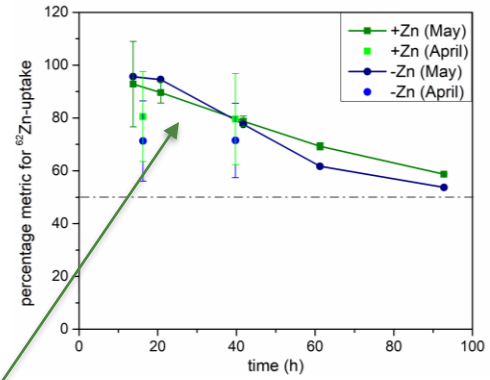
Uptake & Imaging of Garden Cress

- Enhanced uptake from org. Zn-complexes?
  - Cress cultivated for 5 – 7 days in petri dishes
  - Treatment: 14 – 81 kBq ^{62}Zn +/- 50µM DTPA



- Uptake of ^{62}Zn followed by phosphor imaging
  - 8.5 – 93 h post incubation



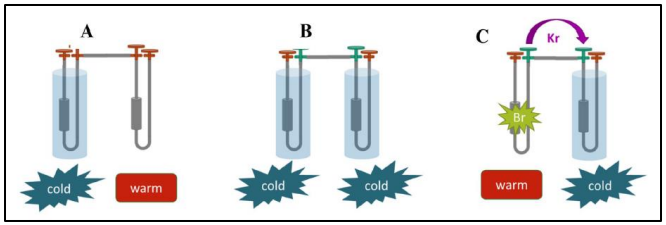


- Trend towards preferred absorption of ligand-free ^{62}Zn (Zn^{2+})

Reference: Domnanich *et al.* (2020)

Isotope Harvesting at NSCL

- Collected activities: µCi – low mCi
  - **Gas phase harvesting of ^{76,77}Kr (Nuclear Med.)**
    - » Successful demonstration of ^{76}Kr/^{76}Br & ^{77}Kr/^{77}Br generator concept
    - » Radionuclidically pure ^{76}Br isolated



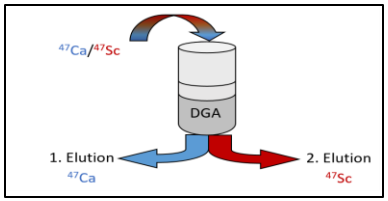


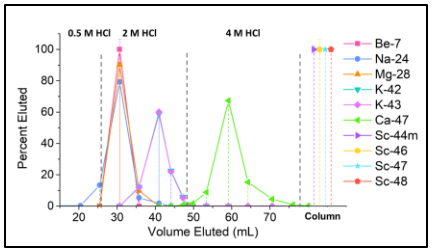
Dr. Hannah Clause

  - **Aqueous harvesting of ^{47}Ca (Nuclear Med.)**
    - » Development of a ^{47}Ca/^{47}Sc generator
    - » First high-intensity harvesting experiment to obtain mCi-levels of pure ^{47}Sc & successful radiolabeling



Dr. Paige Abel





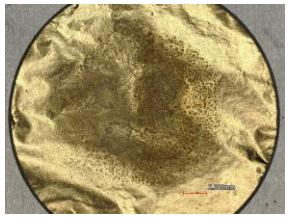
References: Abel *et al.* (2020 a,b), Clause *et al.* (2022)

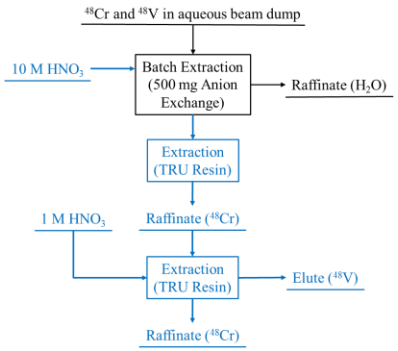
Isotope Harvesting at NSCL & FRIB

- **Aqueous phase harvesting of ^{48}V (Stockpile Stewardship)**
  - » Developed ^{48}V harvesting strategies & ^{48}Cr/^{48}V generator set-up
  - » Prototype V targetry



Dr. Scott Essenmacher





- **Solid phase harvesting of ^{88}Zr and ^{172}Hf (PAC)**
  - » Developed methodologies to isolate ^{88}Zr from irradiated W collectors
  - » & ^{172}Hf from heavy-met alloy (old NSCL beam blocker)
  - » Set-up of ^{172}Hf/^{172}Lu generator



Ms. Samridhi Satija

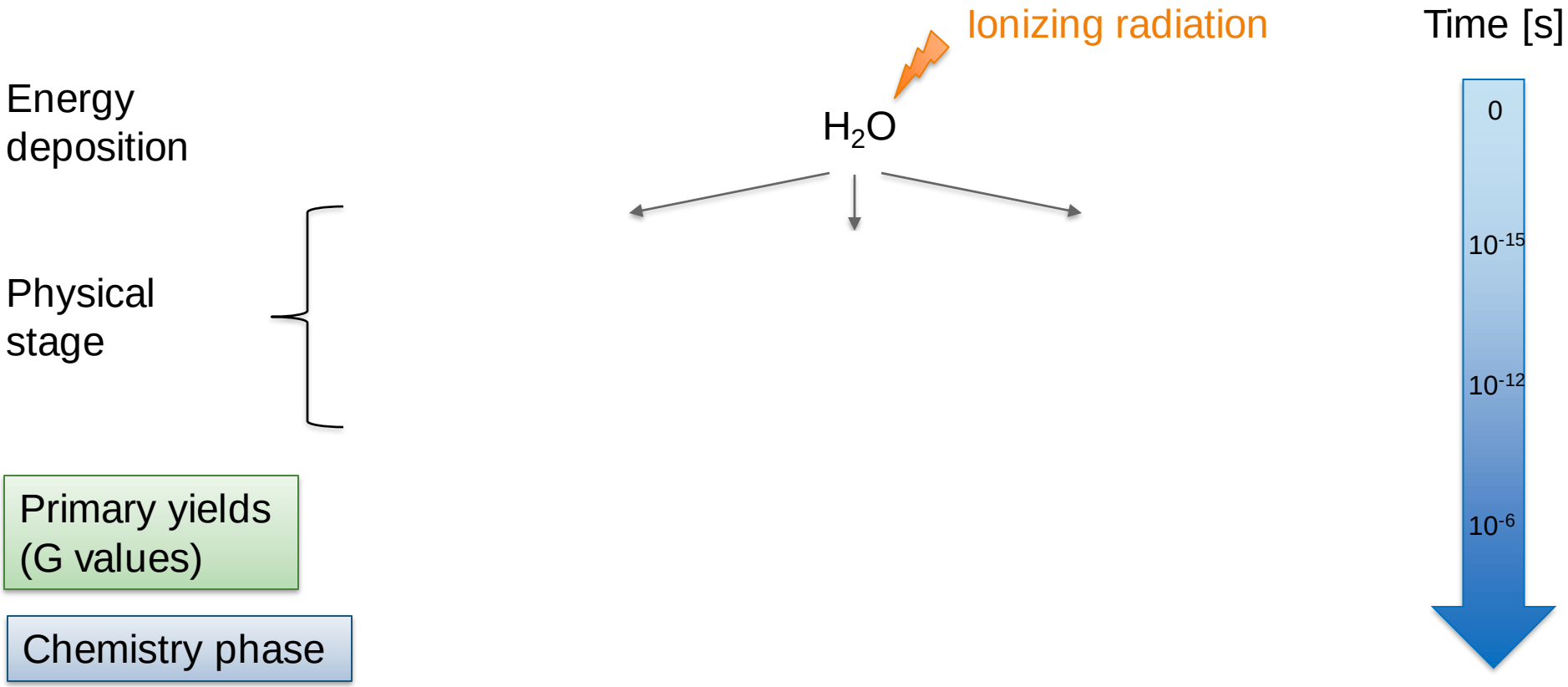


References: Essenmacher *et al.* (2023), Satija *et al.* (2023)

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Radiolysis of Water during Isotope Harvesting



Primary radiolytic yield – Escape yield – G value [molecules/100 eV]

- » Type of radiation, Linear energy transfer, pH of solution
- » Literature data for G(H_2, H_2O_2) acquired at very low beam currents (max. 5 enA)

References: Baldaccino *et al.* (2019), Meesungnoen *et al.* (2005), La Verne (2004)

Why studying Water Radiolysis?

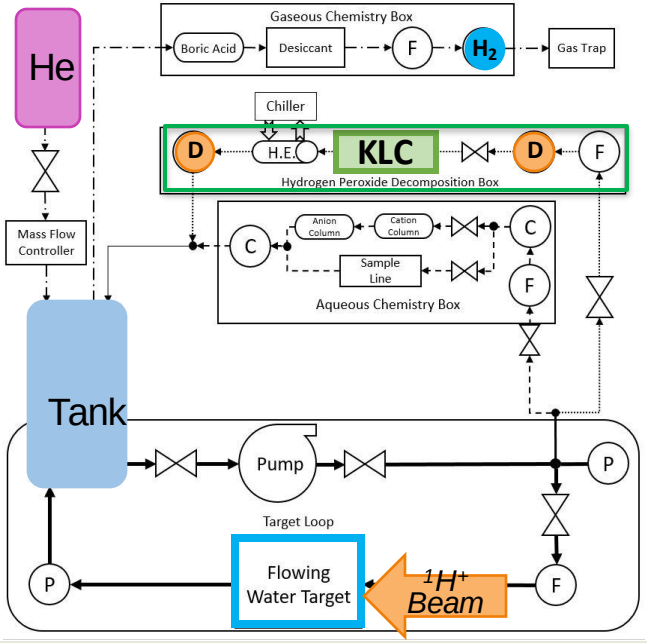
- H_2O_2
  - Corrosive towards equipment
  - Influence on redox environment
- H_2 and O_2
  - Influence on redox environment
  - Harvesting of gaseous radionuclides
- $^1H^+$-beam at University of Wisconsin-Madison
  - Investigate durability of flowing-water target
    - 16 MeV $^1H^+$
    - Intensity: 1 – 34 μA
    - Total t_{irr}: 190 min
      4 run segments

Decomposition of H_2O_2 by catalytic converter (KLC)





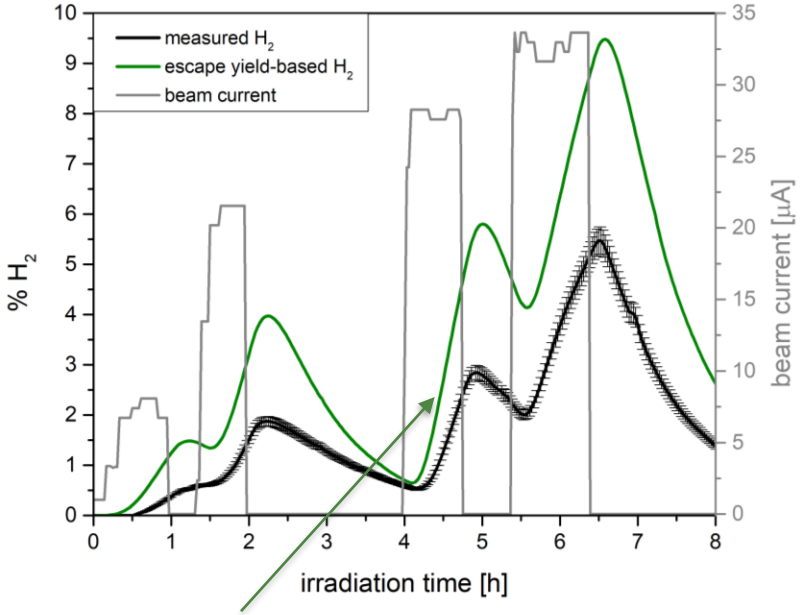
$H_2O_2 \rightarrow H_2O + ½\ O_2$



Reference: Abel *et al.* (2020)

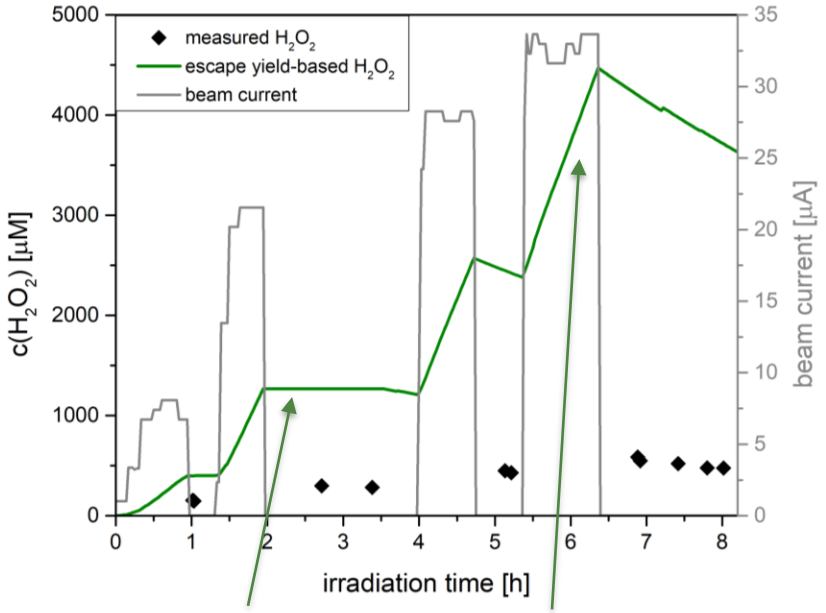
Comparison with Escape Yield Prediction

H_2 (G_0: 0.64 molec./100eV)



~2 times higher % H_2 predicted

H_2O_2 (G_0: 0.74 molec./100eV)



2.5 – 8 times higher c(H_2O_2) predicted

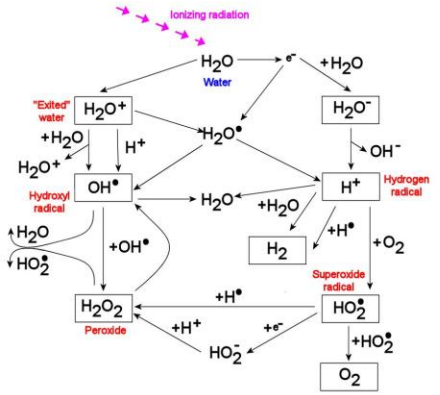
- Similar observation for harvesting experiments at NSCL with ^{48}Ca- and ^{78}Kr-beams at high currents (24 – 695 enA)

For **our setup & irradiation conditions:**
→ **no satisfactory prediction** with the escape yield model

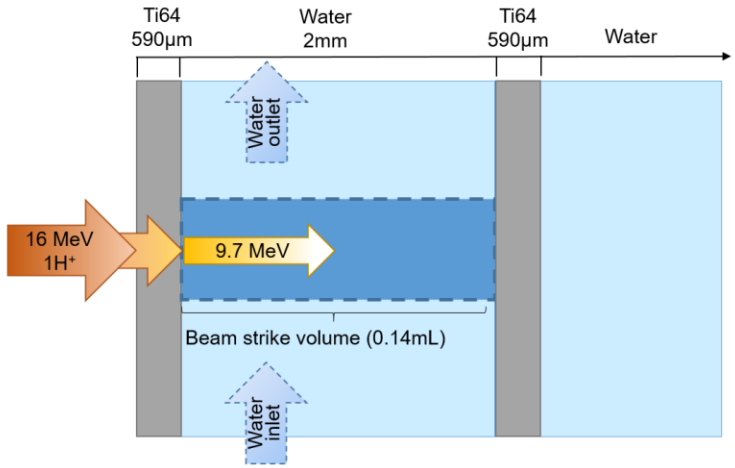
References: Abel *et al.* (2020), Domnanich *et al.* (2022)

Next Step: Homogeneous Kinetic Model

- Development of a model considering the homogeneous kinetics of the chemistry phase
  - 9.7 MeV $^{1}H^{+}$ beam delivered into water phase of target
    - » Escape yield-based formation of primary radiolytic species e^{-}, H, H_2, OH, H_2O_2, HO_2, H^{+}, OH^{-}
    - » Homogeneous reactions of 1st species
    - » Formation of 2nd species: HO_2^{-}, O_2^{-}, O_2, O^{-}, O_3^{-}, O_3, HO_3
    - » Extended life-times of molecular products: H_2, H_2O_2, O_2



- Water flow through flowing-water target
  - » In beam strike volume: 41.7 mL/s

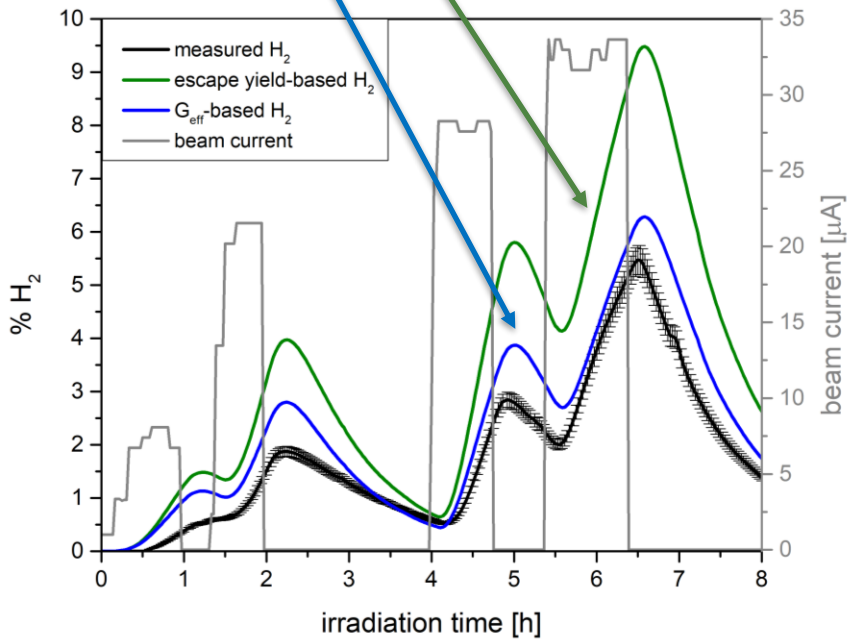


Reference: Domnanich *et al.* (2022)

G_{eff}-based Predictions

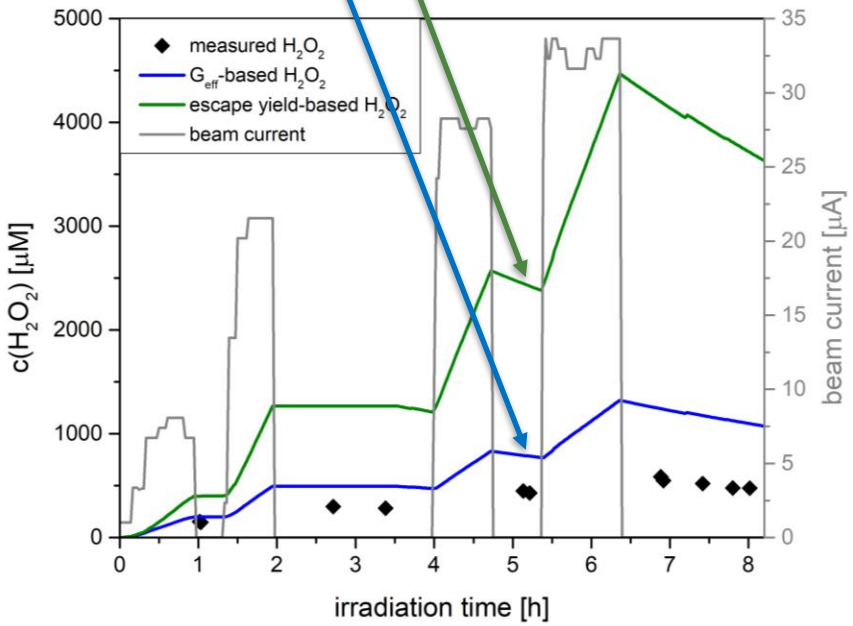
H_2

- Escape yield: ~2-times higher
- G_{eff}-based: ~1.3-times higher



H_2O_2

- Escape yield: 2.5 – 8-times higher
- G_{eff}-based: 1.4 – 2.2-times higher



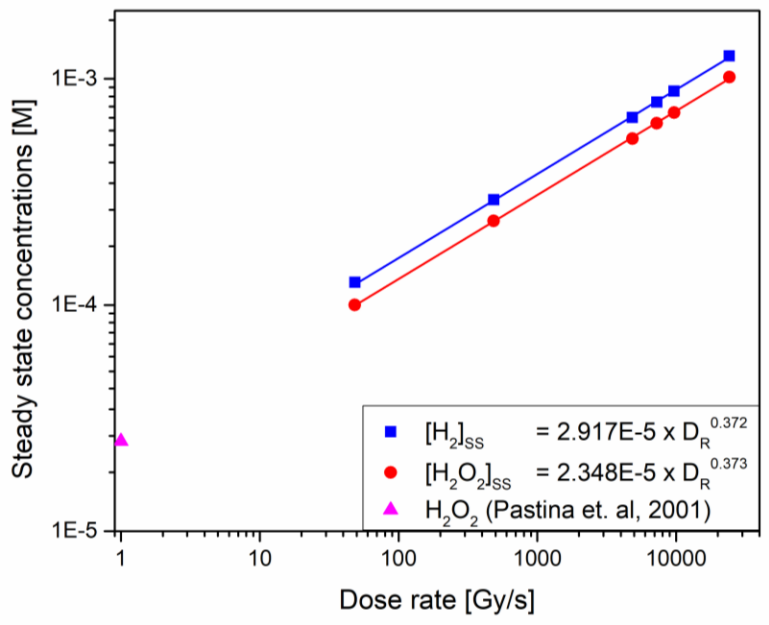
Better approximation with G_{eff}-based predictions BUT still too high in comparison to measured H_2- & H_2O_2-levels.

Reference: Domnanich *et al.* (2022)

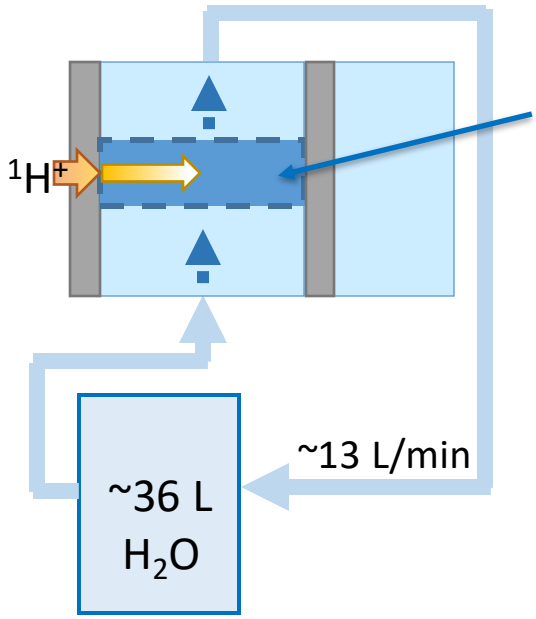
…Considering the Steady State

- Approximation of Steady State (SS) in our system?
  - Current literature: only for closed container with small volume

Dependency on dose rate



SS in our harvesting system



4.3 ms in beam strike volume (0.14 mL)

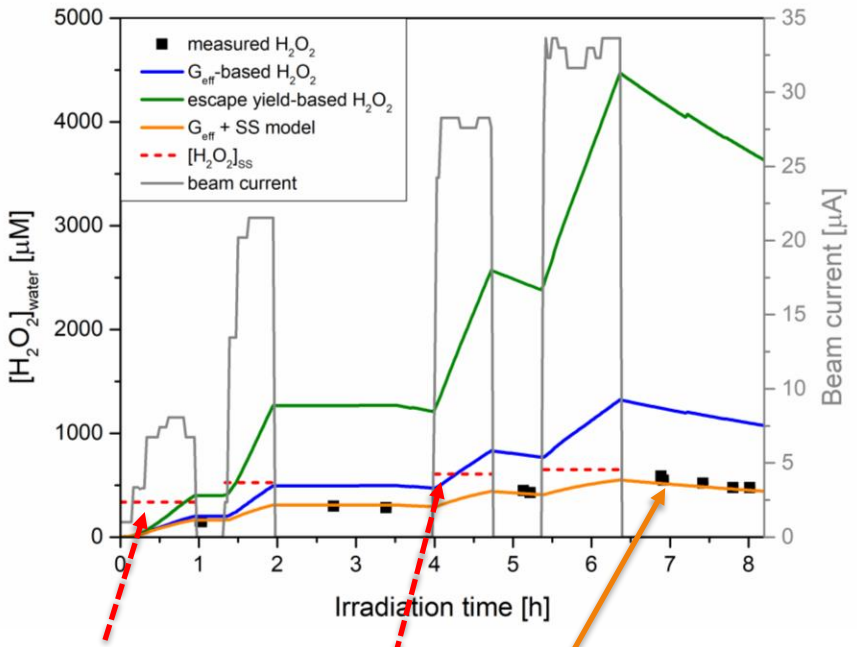
After ~5 ms: onset of SS

- ~16 min until all water passes 1x through entire system
- Fitted time constant k_{time} (2.52E-4 s^{-1})

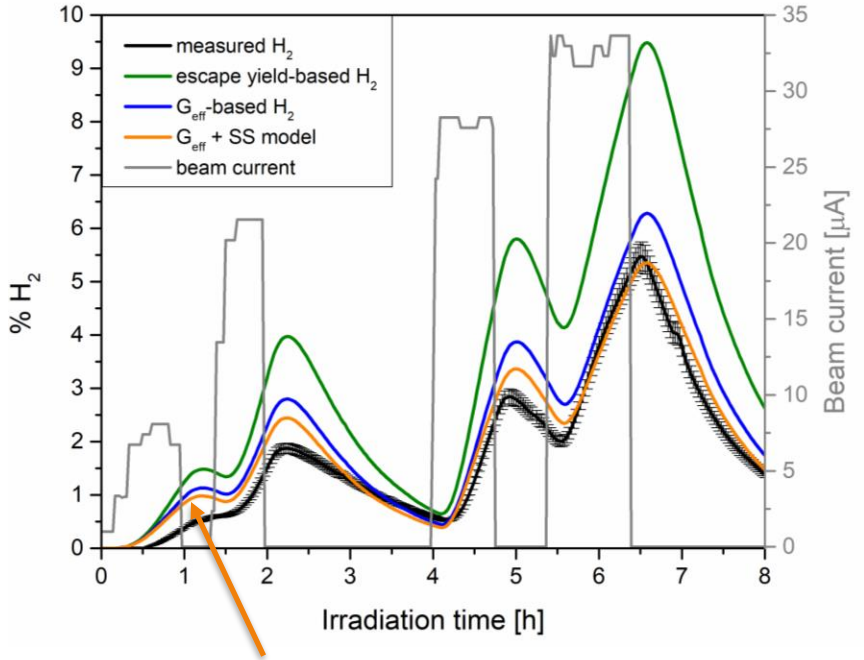
H_2O_2 & H_2 by considering G_{eff} & SS

- Entire system approaches SS

H_2O_2 in water



H_2 at sensor



Calculated $[H_2O_2]_{SS}$ for each irradiation segment

G_{eff}-mediated formation & attenuating effect of established SS

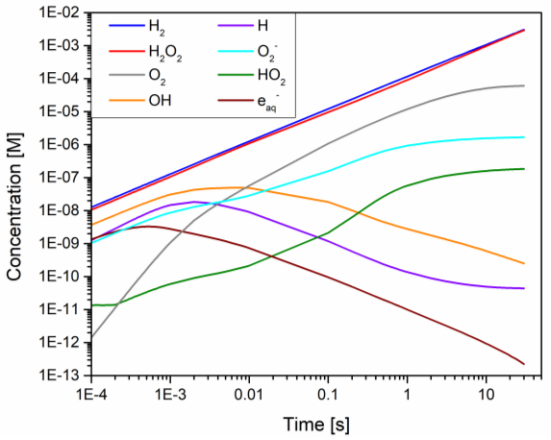
Fitted k_{time} → not yet confirmed experimentally

Reference: Domnanich *et al.* (2022)

Translation towards Heavy Ion Irradiations

- SS-approximation with low LET radiation (X- & γ-rays)
  - High radical yield induces decomposition rxn.

4 µA α-beam (5 MeV), 1 kGy/s



- High LET radiation (heavy ions)
  - Continuous formation of molecular products

Initial beams at FRIB

- Beam power: 10 – 50 kW
- Dose rate f. ^{48}Ca-beam: 2.7 kGy/s
- Mixed radiation field
  - 1st, 2nd (3rd) ion beam particles: light – heavy
  - High energy neutrons, γ-rays
- Options to mitigate radiolysis
  - H_2 addition if sufficient radicals present
  - Alternative: catalytic H_2 & H_2O_2 decomposition

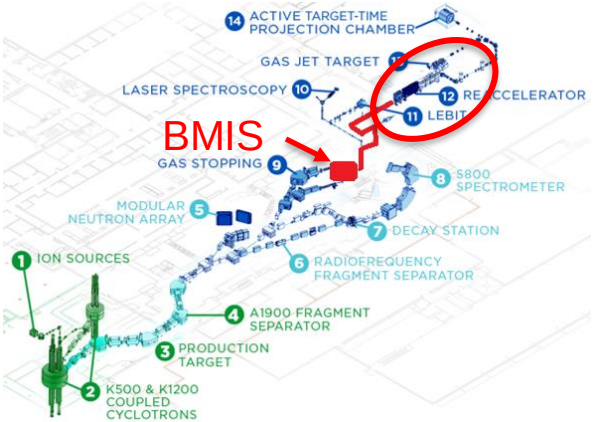
Overall G_{eff}
→ Dependent on beam type & energy

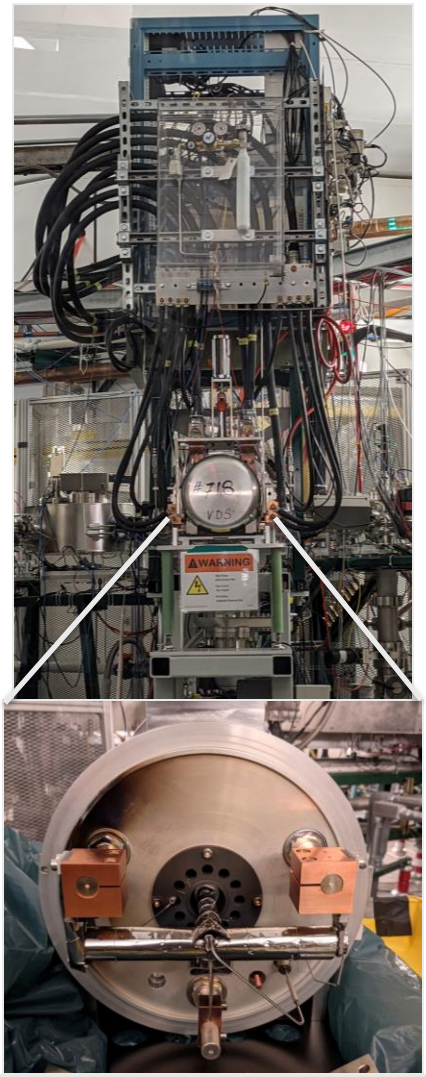
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Preparation of Radioactive Source Samples

- **B**atch-**M**ode **I**on **S**ource allows stand-alone operation of stopped-beam experiments (ReA, LEBIT, BECOLA)
  - Stopped beams from FRIB re-accelerated
  - Radioactive and stable source samples necessary
    - » Into oven → atomized & ionized (mostly +1)



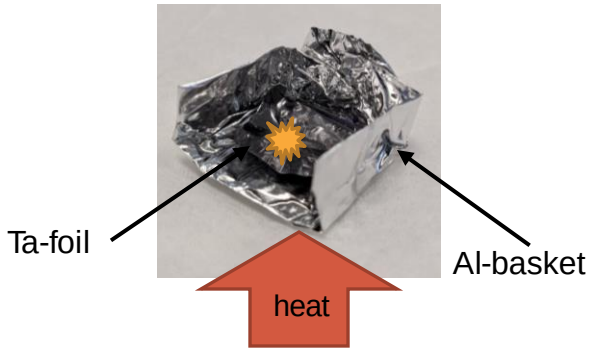


- Oven Temp: ≥ 300 to ~ 1800 ° C

References: https://nscl.msu.edu/public/virtual-tour

Current Sample Source Program

- Sample source preparation program since Sept. 2021
- Requirements for BMIS source sample
  - Volatile at operating Temp. → melting point of compound
  - No isobaric impurities
  - Avoid high matrix mass
- Experimentally
  - Chem. Transformations to produce adequate compounds
    - » ^{7}Be ($t_{1/2}$: 55.2 d): 4 x 0.1 µCi – 4.8 mCi $^{7}Be(NO_3)_2$/ $^{7}BeCl_2$
    - » ^{32}Si ($t_{1/2}$: 153 a): 2 x 10^{15} atoms $Na_x{}^{32}Si_yO_z$
    - » ^{26}Al ($t_{1/2}$: 7.2x10^{5} a): 2 x 10^{15}–10^{16} atoms $^{26}Al_2(SO_4)_3$
    - » ^{73}As ($t_{1/2}$: 80.3 d): 1 x 10^{16} atoms $Ag_3[^{73}AsO_4]$
    - » Stable ^{30}Si, ^{50}Cr, ^{58}Fe: 10^{18}–10^{19} atoms
  - Transfer of radioisotope on Ta-foil



Reference2: Domnanich *et al.* (2023), Sumithrarachchi at al. (2023)

Acknowledgements

- Domnanich group

  Andrew Candia, Suvan Campbell, Monika Fouad

- Isotope harvesting gang @ MSU

  Prof. Gregory Severin, Paige Abel, Jose Blanco, Vlad Bodnar, Hannah Clause, Tracy Edwards, Scott Essenmacher, Alyssa Kalman, Chloe Kleinfeldt, Samridhi Satija, Chirag Vyas, Wes Walker

- Scientists @ NSCL

  Thomas Bauman, Georg Bollen, Yuan Liu, Dave Morrissey, Stefan Schwarz, Matthias Steiner, Chandana Sumithrarachchi, Andreas Stolz, Antonio Villari

- Funding
  - Michigan State University
  - U.S. Department of Energy, Isotope Program

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Thank you for your attention!