FRIB

Water as a Target for Heavy Ion Irradiations

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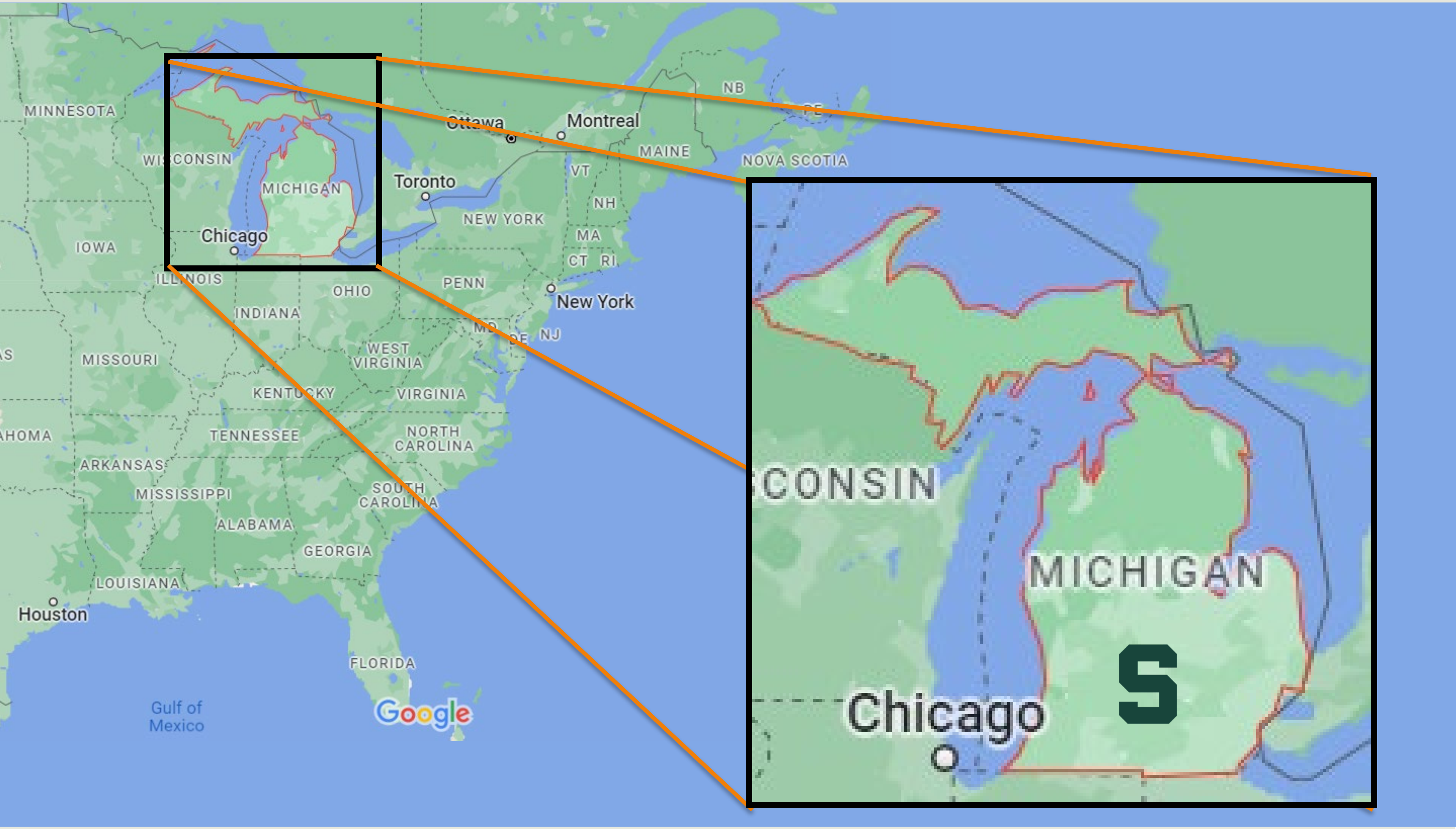
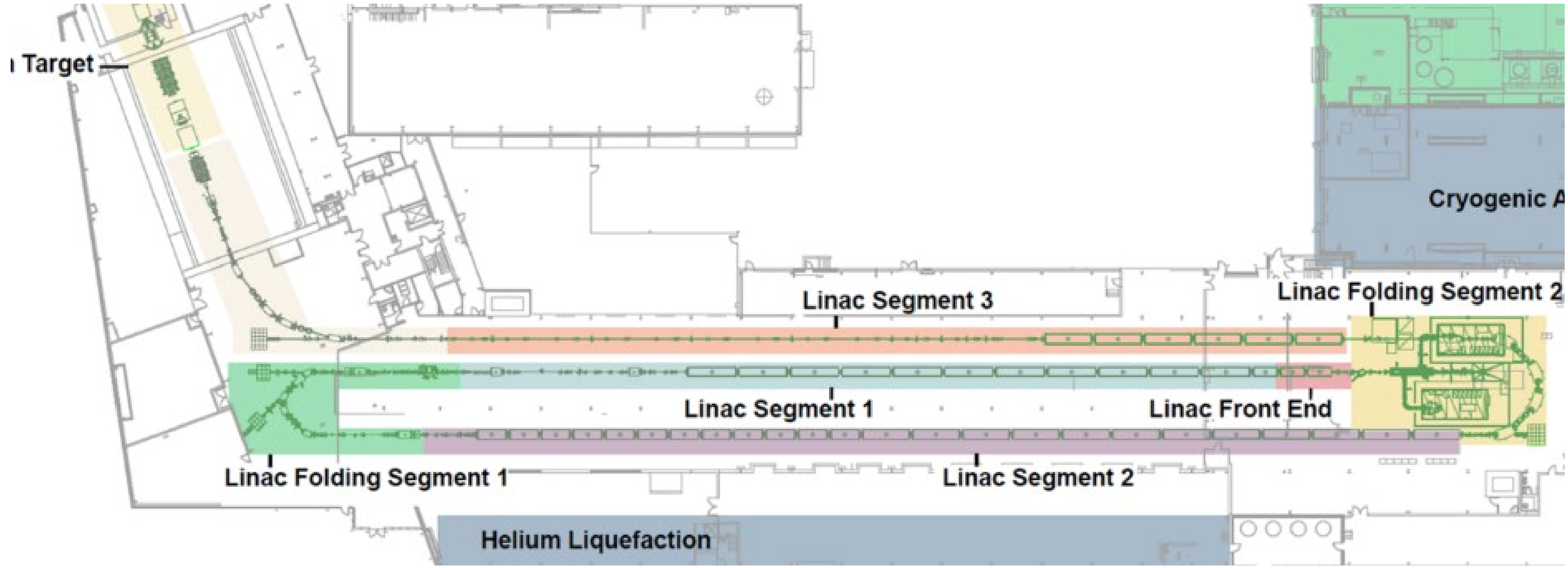
This material is based upon work supported by the U.S. Department of Energy Office of Science Isotope Program

FRIB is a Nuclear Physics Laboratory at Michigan State University in the US



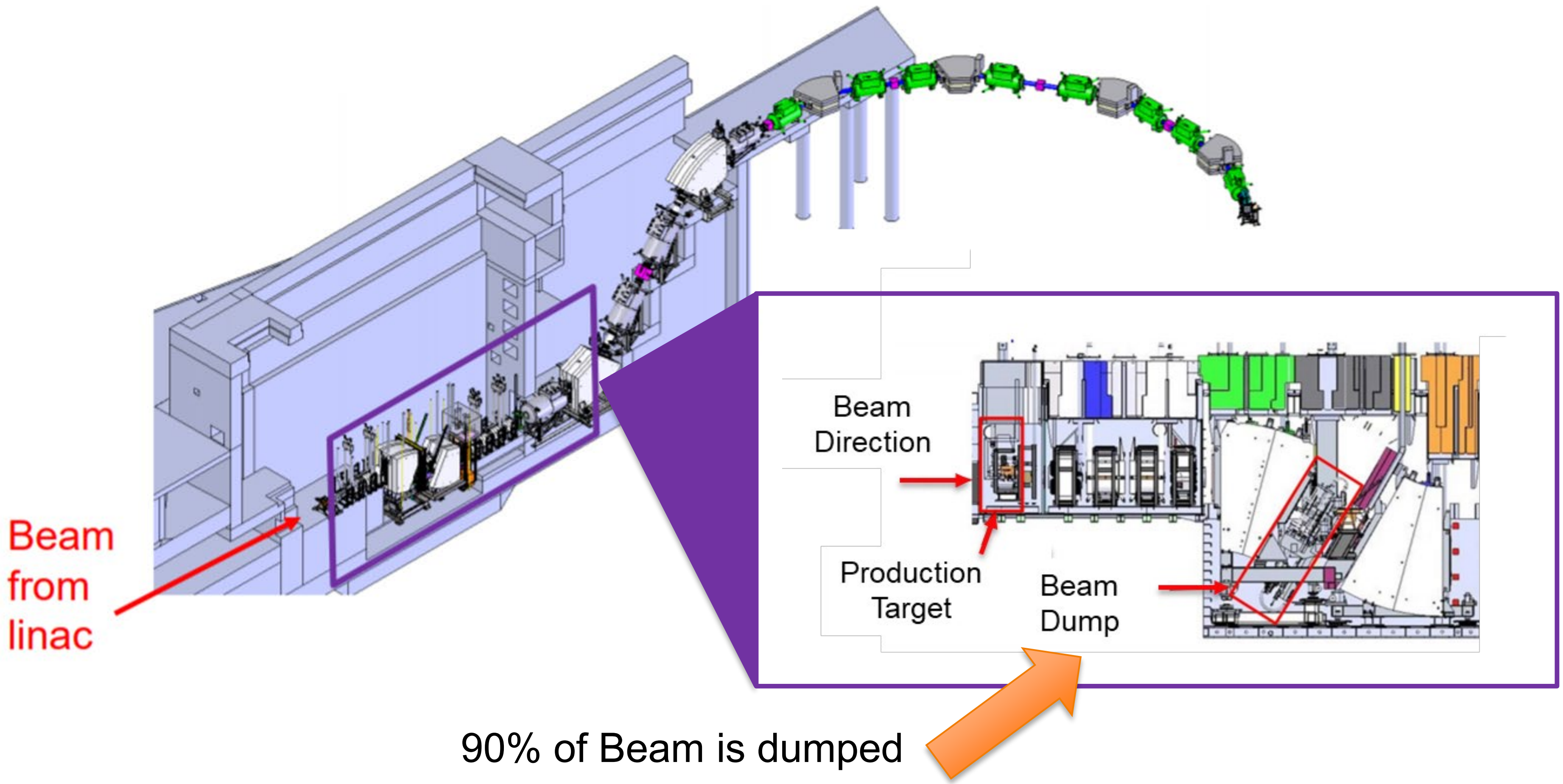
The Heart of FRIB is a Linear Heavy Ion Accelerator



For example:

- $^{48}Ca^{20+}$ is accelerated to > 200 MeV/nucleon, 1-50 μA (up to 400kW)
- The beam hits a thin target and creates fragmentation-product radionuclides inflight
- The fragmentation products are steered to experimental stations downstream

90% of the FRIB Beam is Dumped



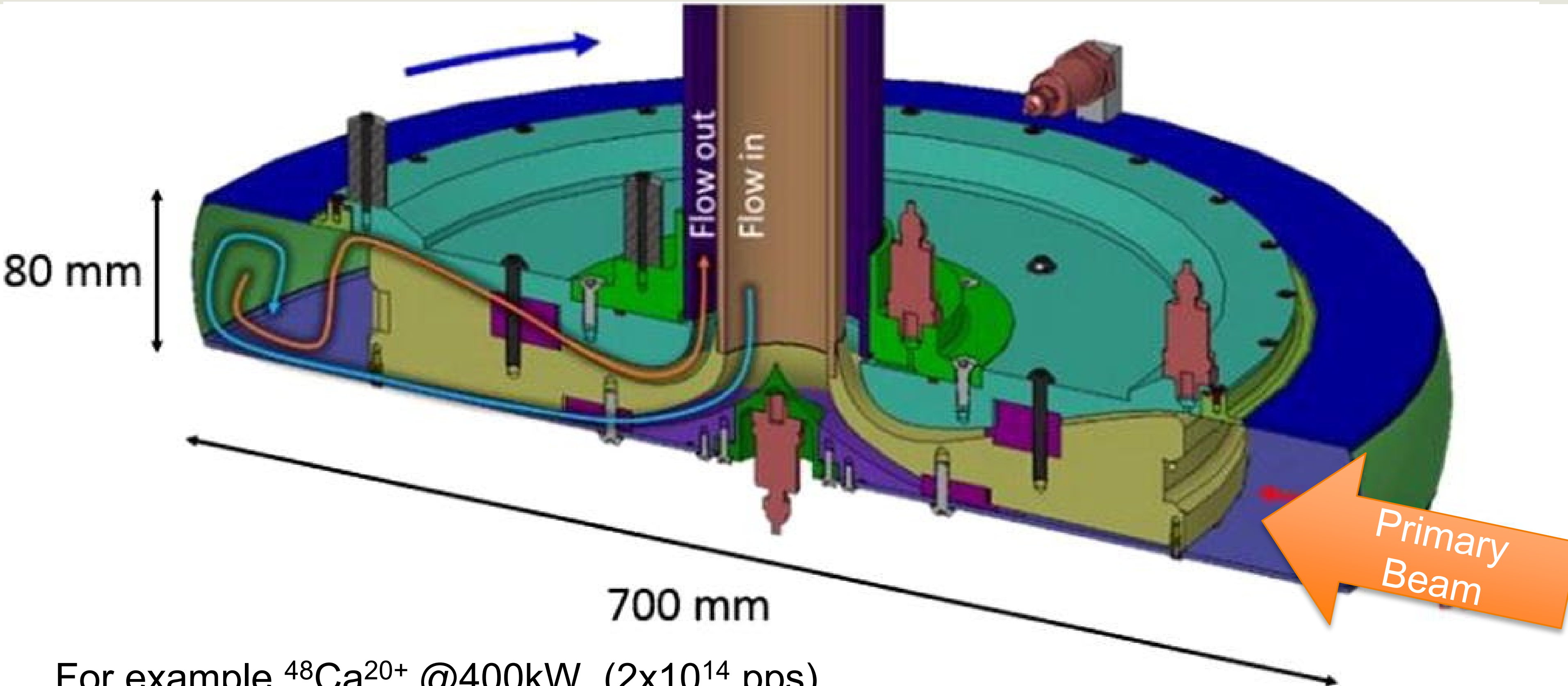
90% of Beam is dumped

Figure adapted from Dali Georgobiani, FRIB Radiation Studies, HPT Workshop, June 2018

What can you do with excess beam (instead of dumping it)?

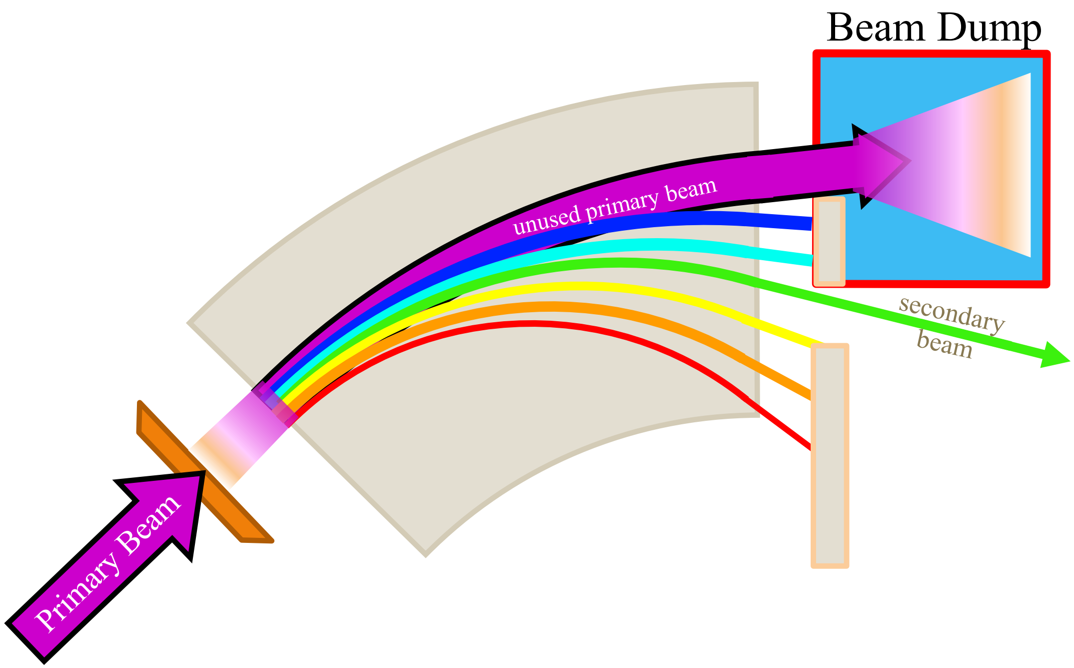
(Photo: Jerry Nickles, UW)

The FRIB water-filled beam dump

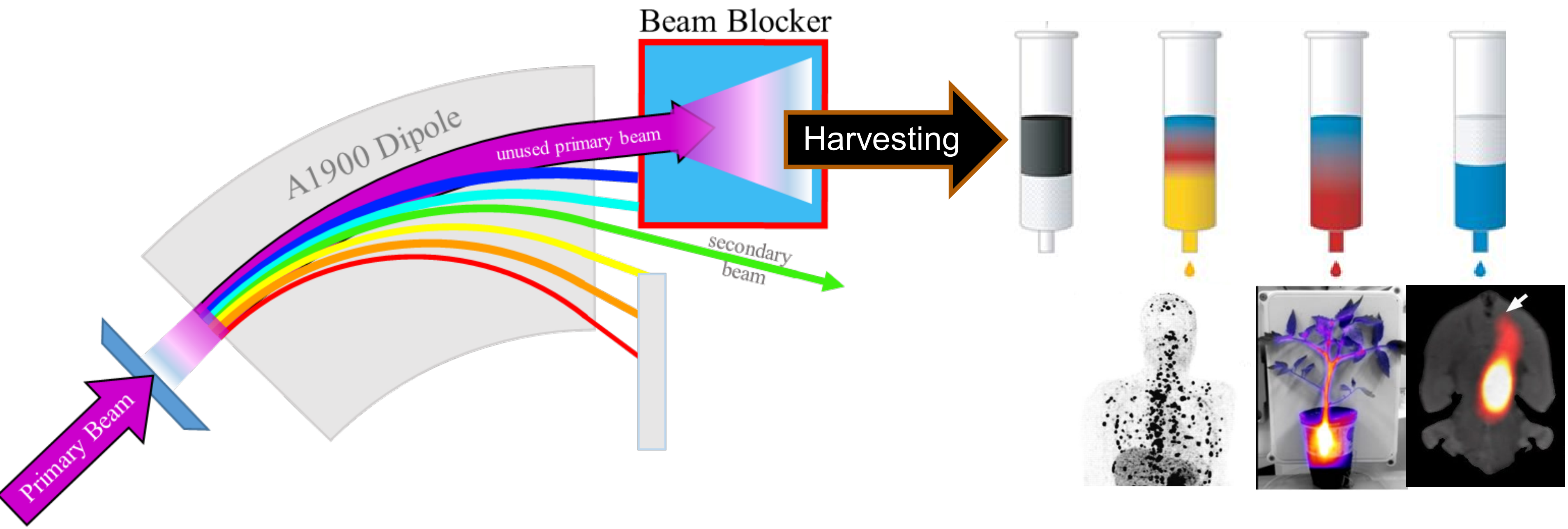


For example $^{48}Ca^{20+}$ @400kW (2x10^{14} pps)

Rare Isotope Production at NSCL/FRIB and Isotope Harvesting



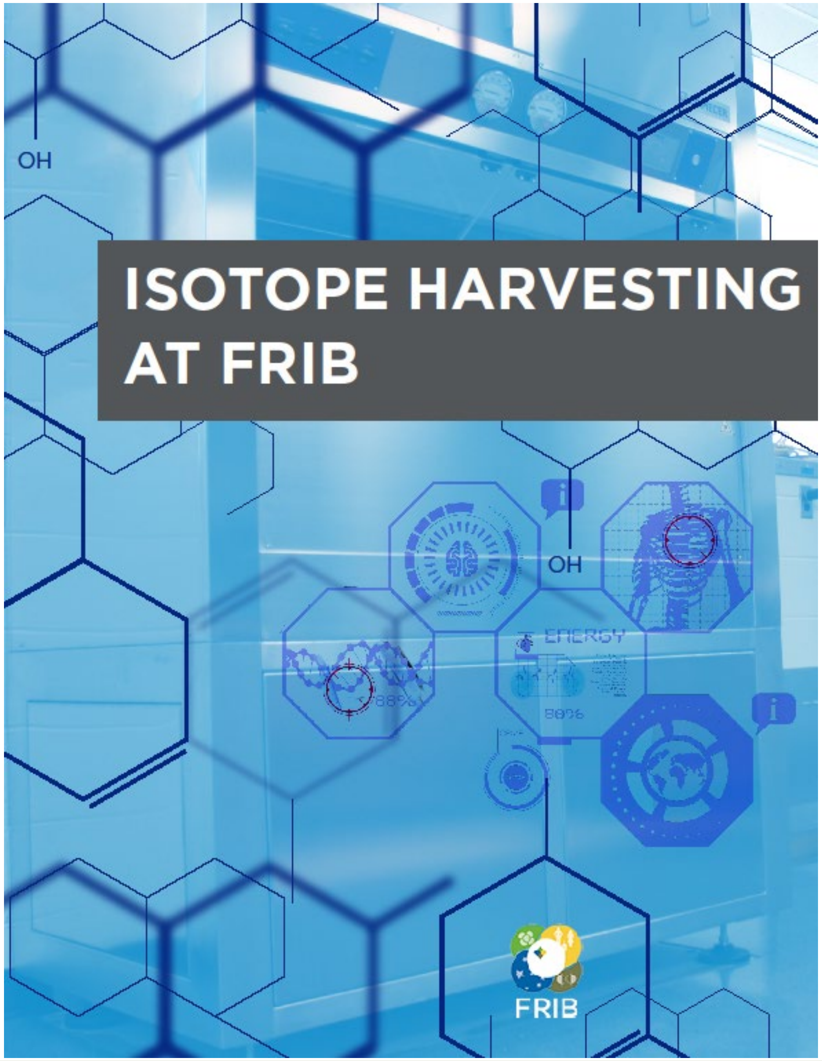
"Harvesting"



(Purify radionuclides using chemistry instead of magnets)

FRIB initial harvesting opportunities

- **^{238}U beam**
  - ^{211}Rn
  - ^{229}Pa
  - ^{225}Ra
- **^{48}Ca beam**
  - ^{47}Ca
  - ^{22}Na
- **^{78}Kr beam**
  - ^{76,77}Kr
  - ^{72,73}Se



Journal of Physics G: Nuclear and Particle Physics

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Isotope harvesting at FRIB: additional opportunities for scientific discovery

https://iopscience.iop.org/article/10.1088/1361-6471/ab26cc

The Challenges of Using Water as a Target

- Beam + Water = Radiolysis
  - Radiolysis chemistry affects harvesting (oxidation state, materials lifetimes)
- Extremely Dilute Analyte
  - We need to collect a small amount of radionuclides out of a big amount of water, gas, and solids
- Many nuclear reactions and products
  - A Short ^{48}Ca irradiation at NSCL created 16 quantifiable radionuclides

Radiolysis of Water: see Kathi's Talk

Ionizing radiation

Time [s]

Energy deposition

H_2O

Physical stage

H_2O^* H_2O^+ e^-

OH H_3O^+ OH^- H H_2 e^-_{aq}

Primary yields (G values)

H OH HO_2 e^-_{aq} H_3O^+ OH^- H_2 H_2O_2

Chemistry phase

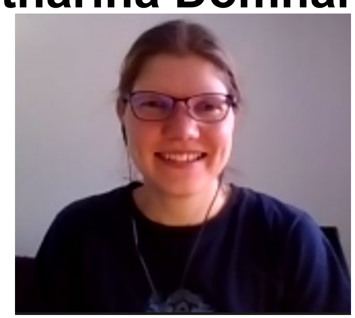
0

10^{-15}

10^{-12}

10^{-6}

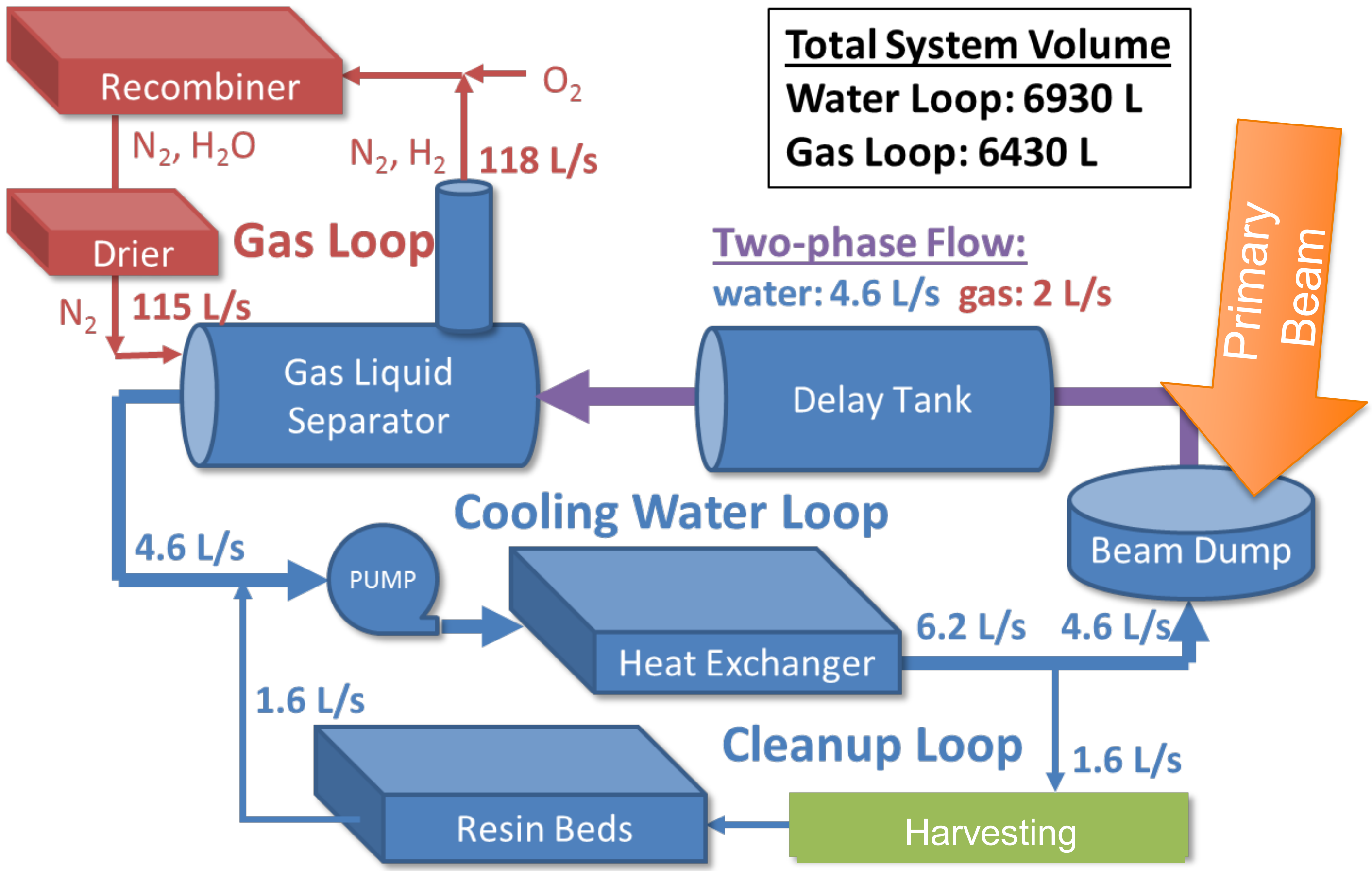
Katharina Domnanich



Reference: Baldaccino *et al.* (2019)

Dilute Samples (orders of magnitude)

- FRIB beam dump cooling system:
  - 10^4 L of water moving at 10^1 L per second
- Beam at full power is few x 10^{14} particles per second
  - ~100 pmol / sec
- Production rates for interesting radionuclides
  - 10^{-3} per incoming beam particle
- After 1 day of irradiation we have 10^4 pmol of analyte in 10^4 L of water
  - 1 pM



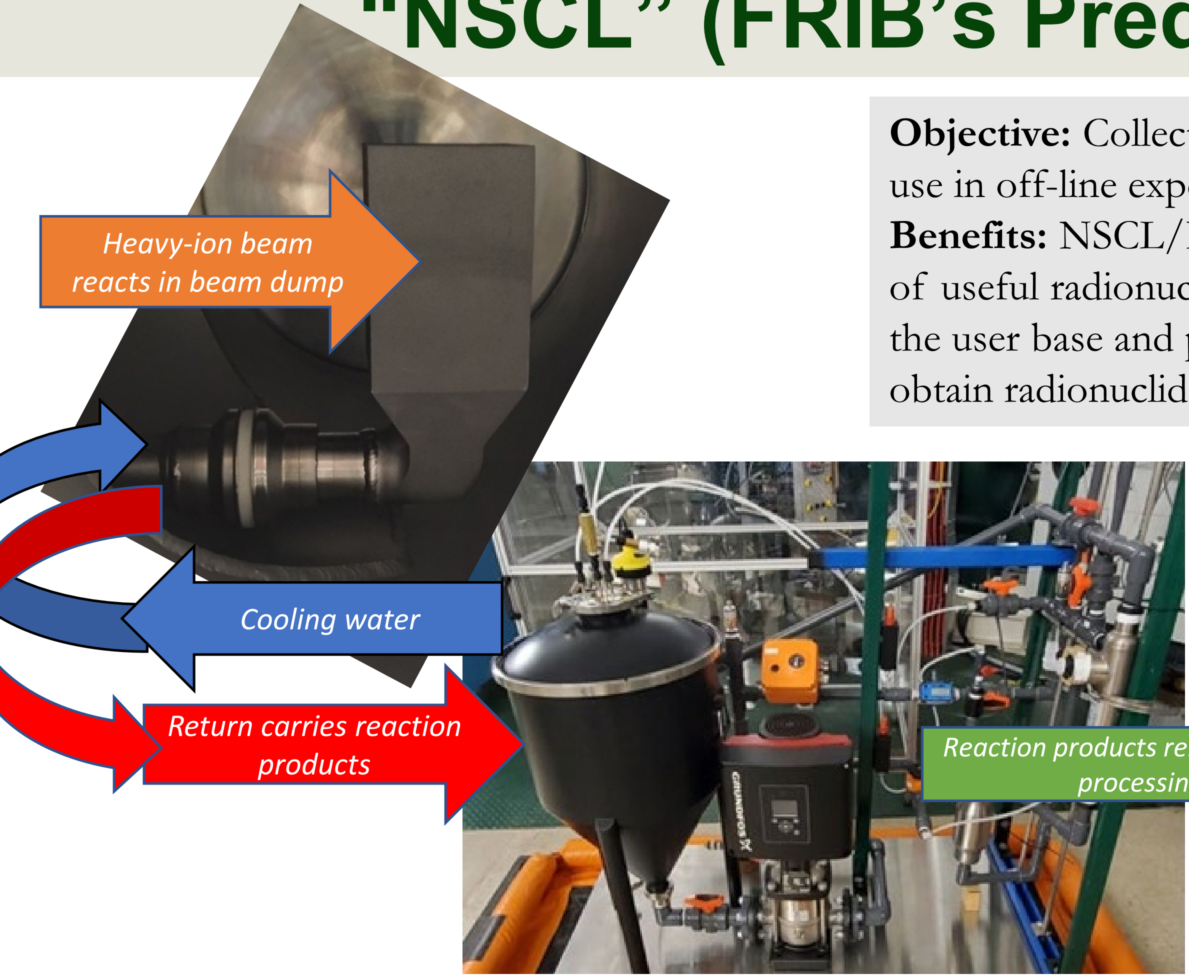
Lots of different radionuclides (many of them are even *isotopes*)

- Number of atoms of each radionuclide after 1 year irradiation with 48Ca at full power and 1 hour of decay time (82 of them).

H	3	6.3527E+16	P	32	1.3709E+14	K	45	1.2913E+10	Cr	48	1.1633E+09
Be	7	2.0295E+15	P	33	2.4382E+14	Ca	41	4.4684E+14	Cr	49	6.9912E+07
Be	10	3.0339E+15	S	35	7.6601E+14	Ca	45	3.3508E+15	Cr	51	9.5152E+11
C	11	8.3518E+10	S	37	1.4568E+06	Ca	47	1.3929E+14	Cr	56	1.0048E+03
C	14	4.9262E+15	S	38	1.2860E+11	Ca	49	1.2435E+04	Mn	51	2.9846E+07
N	13	2.9783E+09	Cl	34	3.3612E+01	Sc	43	4.4011E+10	Mn	52	3.9663E+10
O	15	2.7865E+02	Cl	34m	9.5043E+04	Sc	44	1.5939E+11	Mn	54	4.4063E+12
F	18	2.3846E+11	Cl	36	2.5272E+15	Sc	44m	3.9891E+06	Mn	56	4.2795E+08
Ne	24	4.8824E+03	Cl	38	8.3869E+10	Sc	45m	2.0263E+01	Fe	53	3.4439E+04
Na	22	7.2429E+14	Cl	39	9.2883E+10	Sc	46	2.2702E+14	Fe	55	3.4221E+12
Na	24	2.1228E+12	Ar	37	1.3631E+14	Sc	47	1.1699E+14	Fe	59	2.8973E+10
Na	24m	4.8636E-01	Ar	39	3.4234E+15	Sc	48	2.9319E+12	Co	55	2.7141E+08
Mg	27	1.6953E+08	Ar	41	3.9803E+11	Sc	49	8.7059E+07	Co	56	1.4357E+11
Mg	28	6.4270E+11	Ar	42	1.8127E+15	Ti	44	5.6037E+12	Co	57	1.1014E+12
Al	26	6.4307E+14	Ar	43	6.1738E+06	Ti	45	3.9499E+09	Co	58	6.0730E+11
Al	28	1.1501E+09	Ar	44	8.5279E+08	Ti	51	3.6622E+03	Co	60	3.8073E+11
Al	29	3.2604E+07	K	38	1.1017E+07	V	47	5.4104E+07	Co	61	1.3424E+06
Si	31	3.7319E+11	K	42	5.5279E+12	V	48	4.7153E+11	Ni	56	1.7450E+08
Si	32	5.8209E+14	K	43	1.5616E+13	V	49	8.6685E+12	Ni	57	1.7296E+08
P	30	2.3370E+02	K	44	3.8731E+10	V	52	2.6685E+02	Cu	62	1.8972E+05
									Zn	62	1.0549E+07

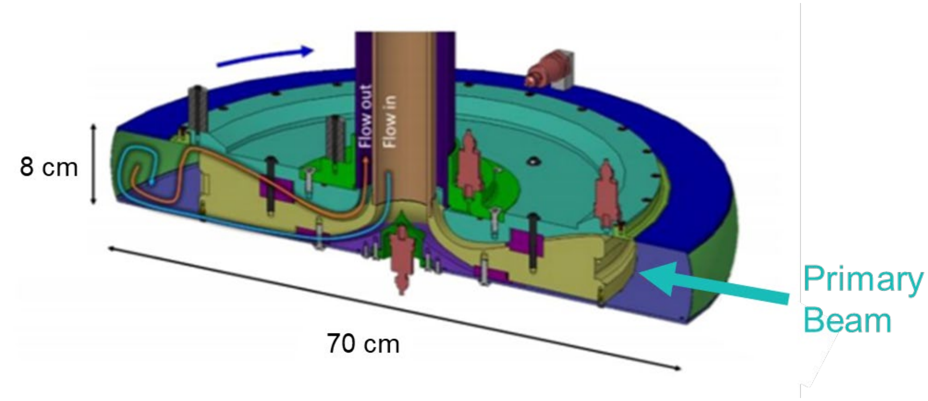
The Harvesting Process was Tested at "NSCL" (FRIB's Predecessor)

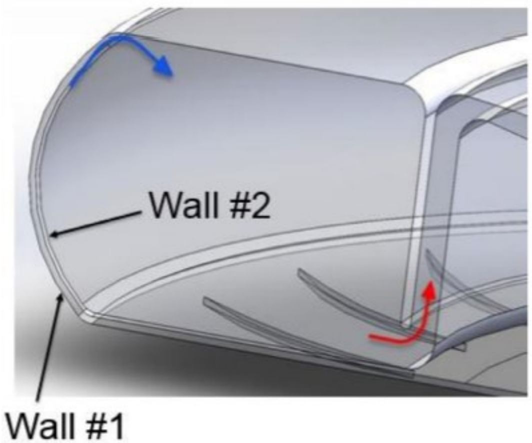
Objective: Collect by-product radionuclides for use in off-line experiments.
Benefits: NSCL/FRIB produce a vast number of useful radionuclides. Harvesting will expand the user base and provide otherwise difficult-to-obtain radionuclides.



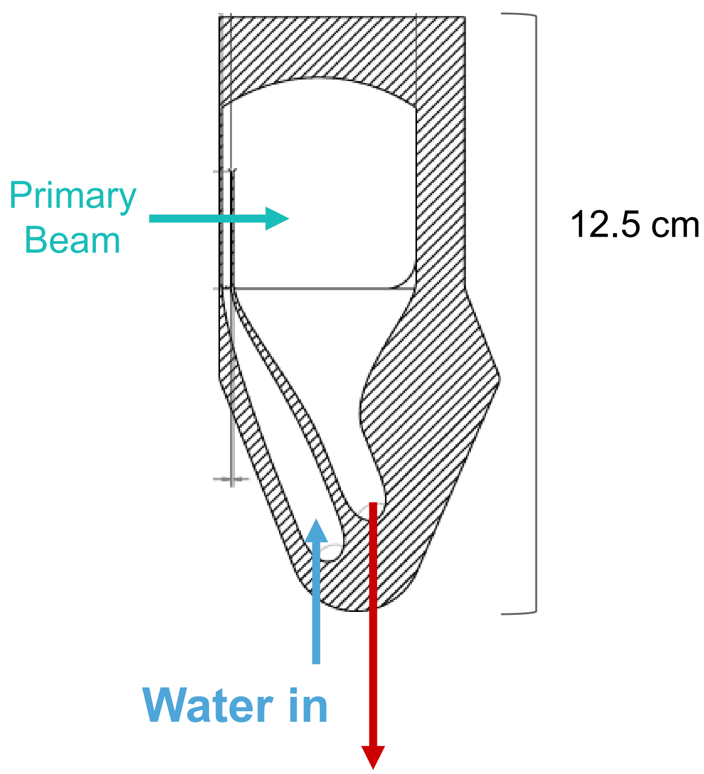
Targetry: Miniature Beam Dump

FRIB Beam Dump





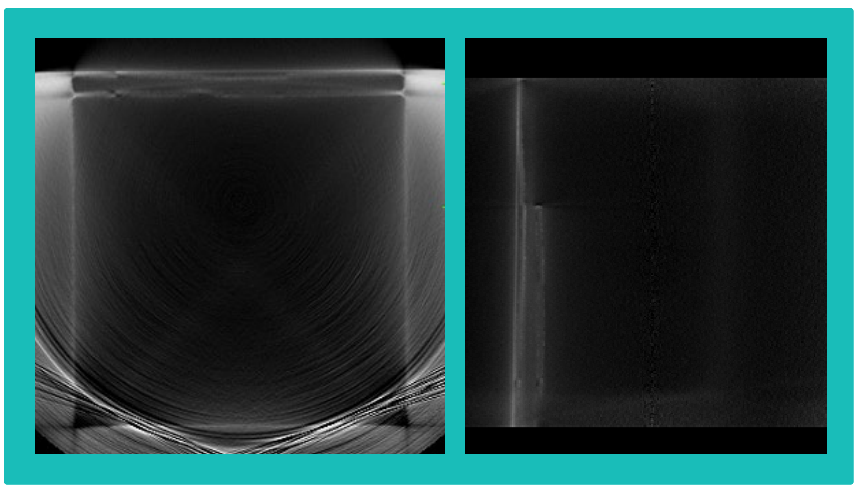
Prototype Beam Blocker

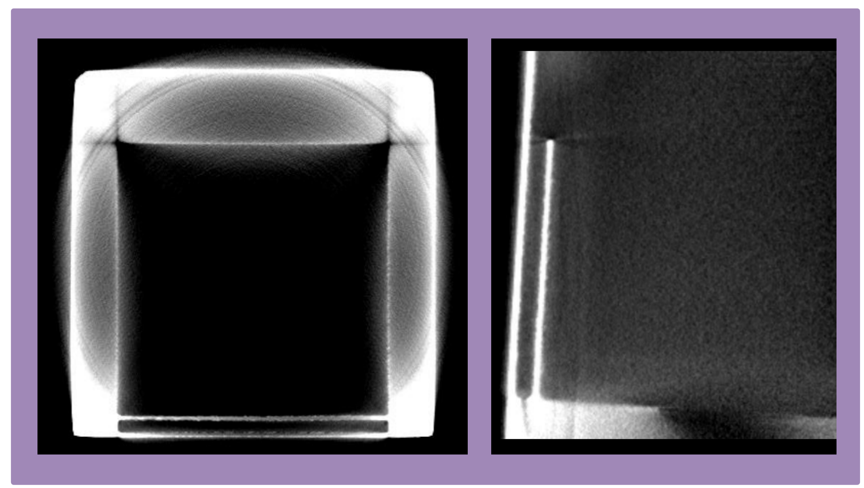


Avilov, M; et.al. Thermal, mechanical and fluid flow aspects of the high power beam dump for FRIB. *Nucl. Instrum. Methods Phys. Res. B*, **2016**, *376*, 24-27.

Production of Target

- 3D printed
  - Ti64 alloy powder (EOS Ti64)
  - Direct metal laser sintering (DMLS)
- CT image of the product
  - Measured thicknesses of thin walls
  - Looked for bubbles in the walls
- Testing the target:
  - Irradiate with high beam current
  - Look for evidence of degradation

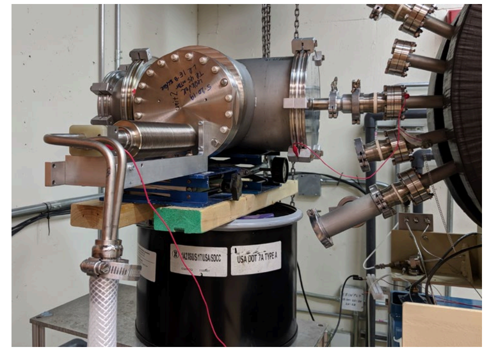


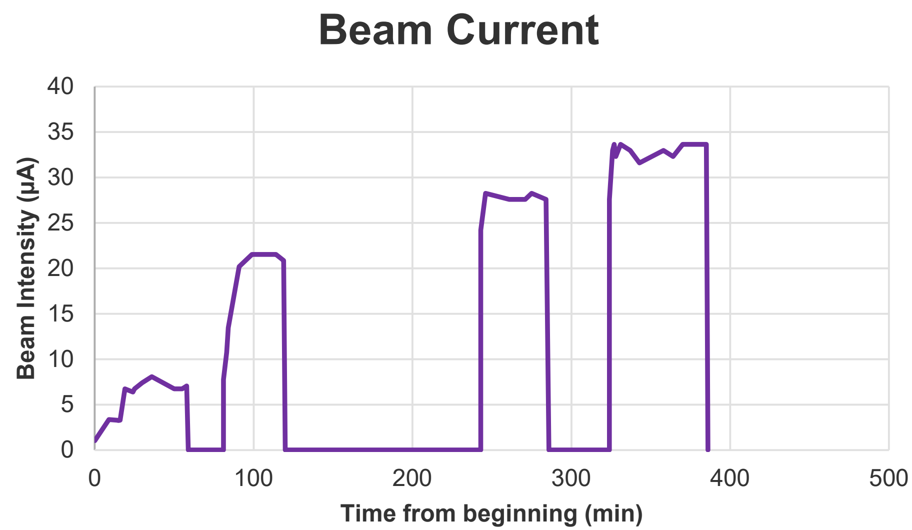


Proton Irradiation Experiment

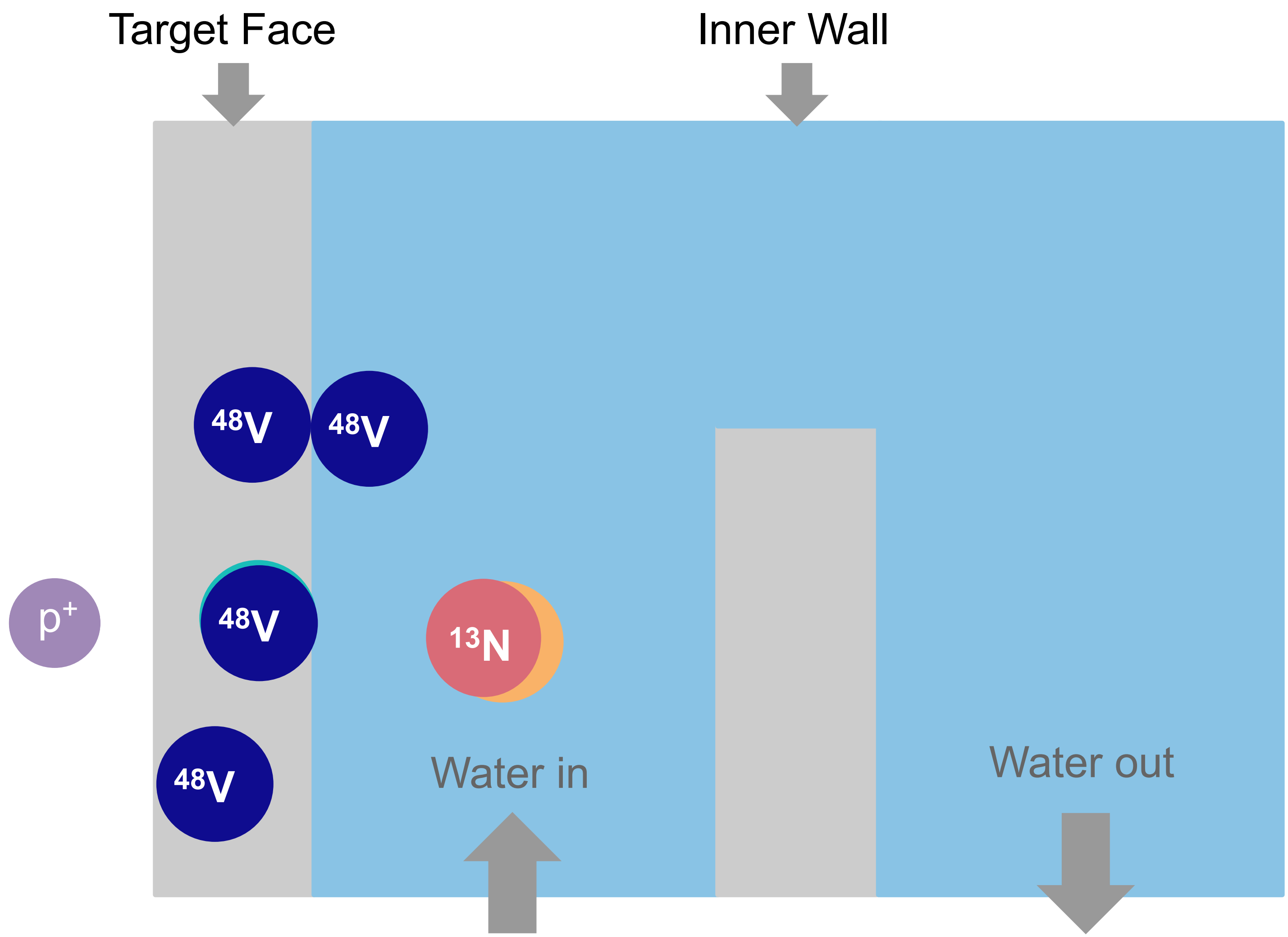
- 16 MeV protons from University of Wisconsin Cyclotron Lab
  - 4 irradiation periods: 6, 19, 28, 33 μA
- Measurements
  - Activity of ^{48}V and ^{51}Cr in system water after each period
  - Total activity of ^{48}V and ^{51}Cr in target face
- One complication: nuclear recoil
  - Forward momentum transfer
  - Estimated activity that left the window

Alexander, J.M.; Sisson, D.H. Recoil Range Evidence for the Compound-Nucleus Mechanism in Reactions between Complex Nuclei. *Phys. Rev.* **1962**, *128*, 2288.





Radiation Accelerated Corrosion? Proton Irradiation Test



Water:

$^{16}O(p,\alpha)^{13}N$

$^{18}O(p,n)^{18}F$

Target Face:

$^{48}Ti(p,n)^{48}V$

$^{51}V(p,n)^{51}Cr$

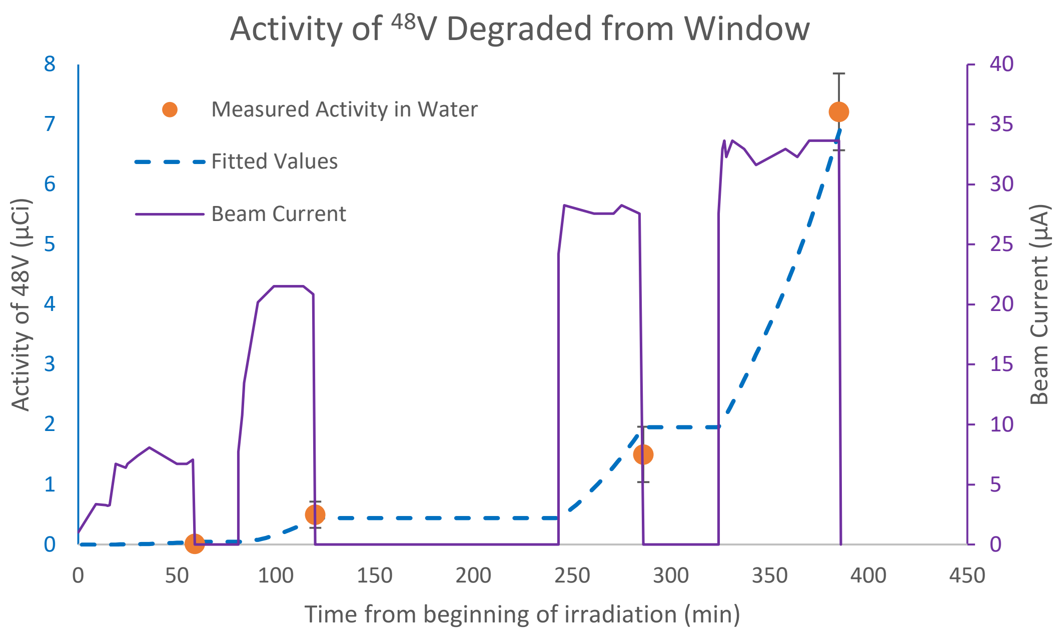
Quantifying Target Degradation

- Basic model:
  - Small time steps
  - Accumulation of ^{48}V in target face
  - Degradation of ^{48}V into the water
    - » Dependence on beam current and activity in target face
    - » Remove activity from nuclear recoil
- Rate: 1.47E-6 μm/(μA*s)

  (X^2 = 1.4)

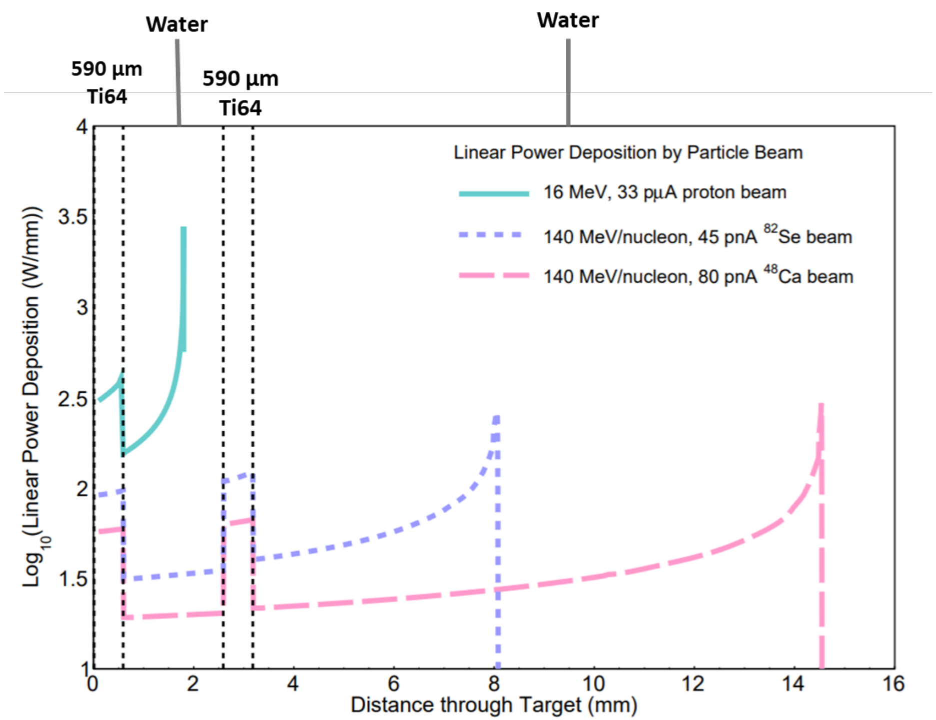
Is this a sufficient model?

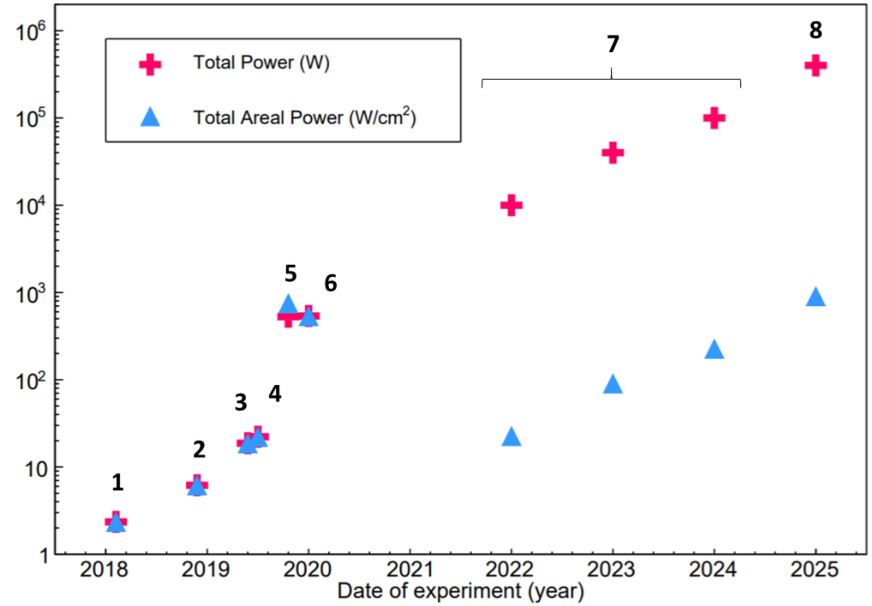
How does this apply to NSCL and FRIB beams?



One Way to Extrapolate

- Scale degradation rate by areal power deposition:
  - Worst Case Scenario for FRIB's dump = 10% degradation after 4000h ^{238}U beam at 400kW.





Summary of Experimental Progress

August 2019: High power proton irradiation

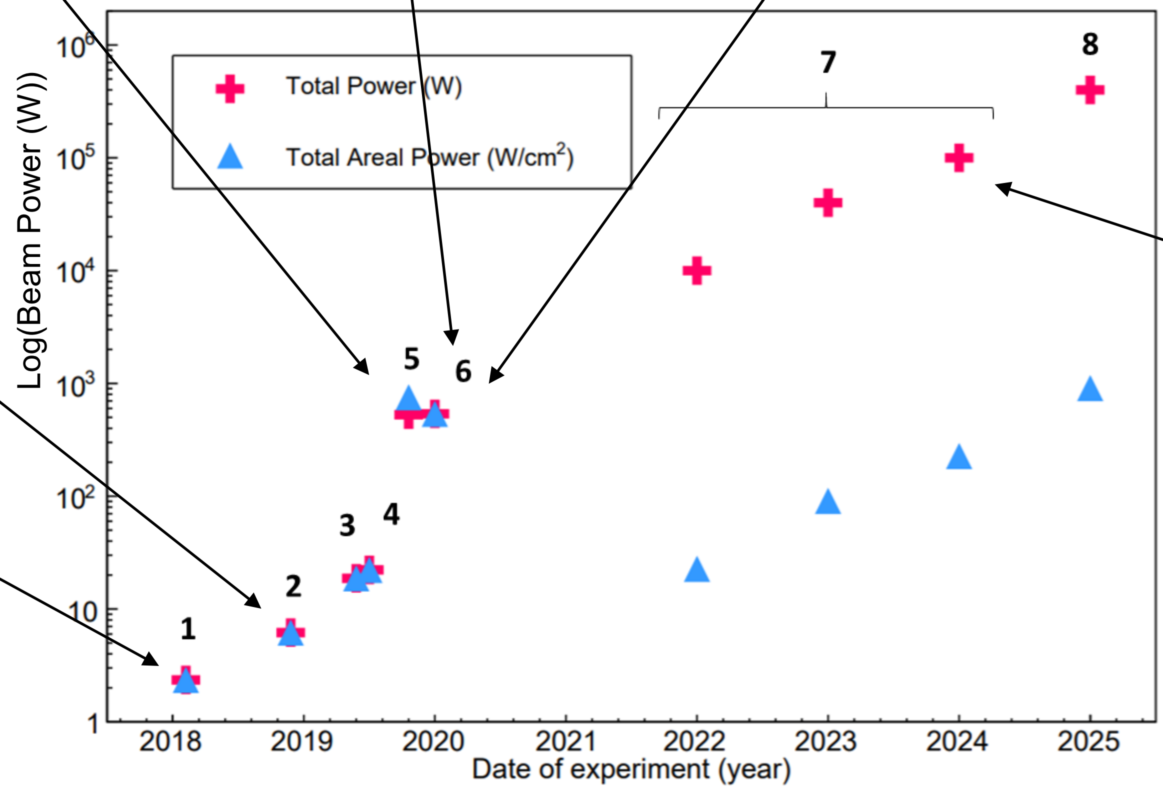
December 2019: Higher power ^{48}Ca beam experiment in beam blocker position

January 2020: Full beam power ^{48}Ca beam experiment (8 hours)

December 2018: First ^{48}Ca beam experiment

February 2018: First flowing-water and primary beam experiment.

2022-2025: FRIB ramp up and full beam power

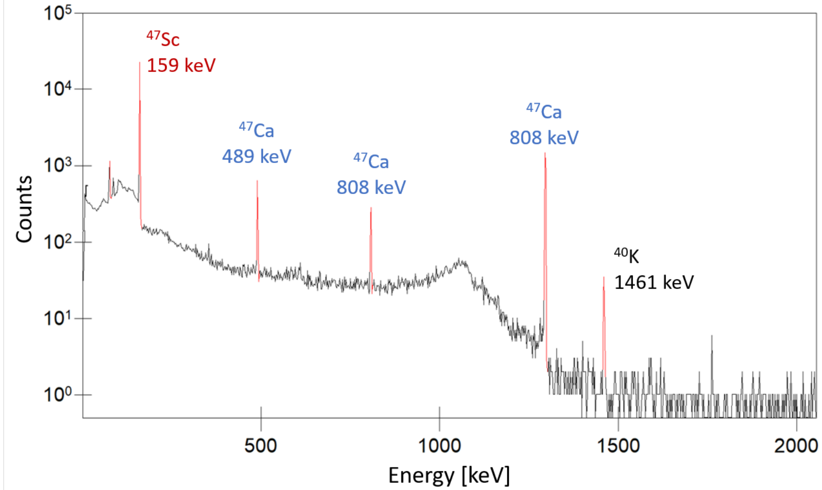


Paige Abel

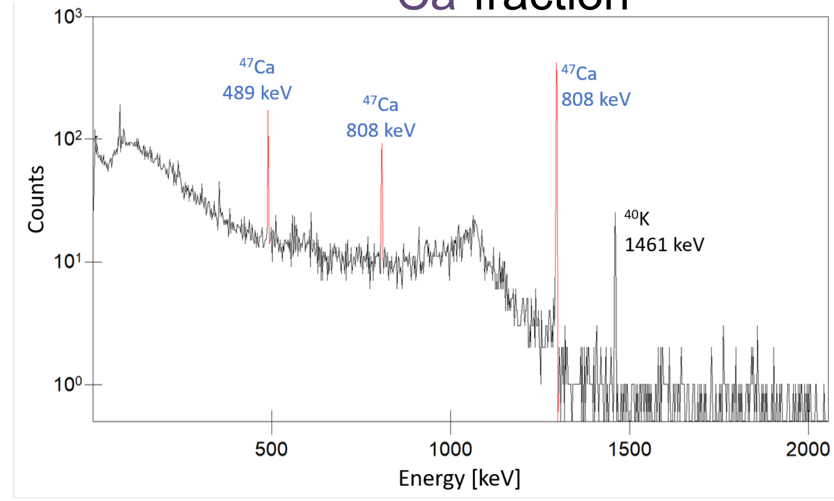
Calcium-48 Beam -- Production

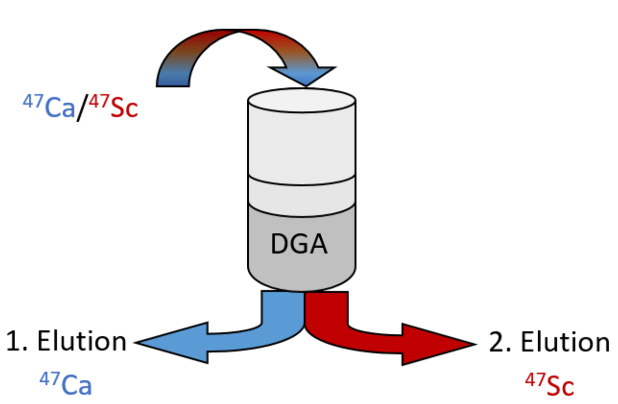
	Percent Conversion	Projected Activity at NSCL (GBq)	
		After 5-day Irradiation	After 24-hour cool down
Na-24	0.084(2)	0.378(7)	0.125(2)
Mg-27	0.0311(8)	0.140(3)	-
Mg-28	0.0187(7)	0.082(3)	0.037(1)
S-38	0.0129(8)	0.058(3)	1.7(1)E-4
Cl-34m	0.00039(5)	0.0177(6)	-
Cl-38	0.116(5)	0.58(4)	-
Cl-39	0.066(2)	0.297(9)	-
K-42	0.343(7)	1.54(3)	0.402(8)
K-43	0.422(3)	1.85(1)	0.878(6)
K-44	0.26(4)	1.2(2)	-
K-45	0.16(1)	0.74(5)	-
Ca-47	1.69(2)	4.05(4)	3.48(3)
Sc-44m	0.482(8)	0.164(2)	0.124(2)
Sc-46	0.52(1)	0.095(2)	0.094(2)
Sc-47	0.41(2)	3.2(3)	3.5(4)
Sc-48	0.220(2)	0.841(9)	0.580(6)

$^{47}Ca/^{47}Sc$ on column

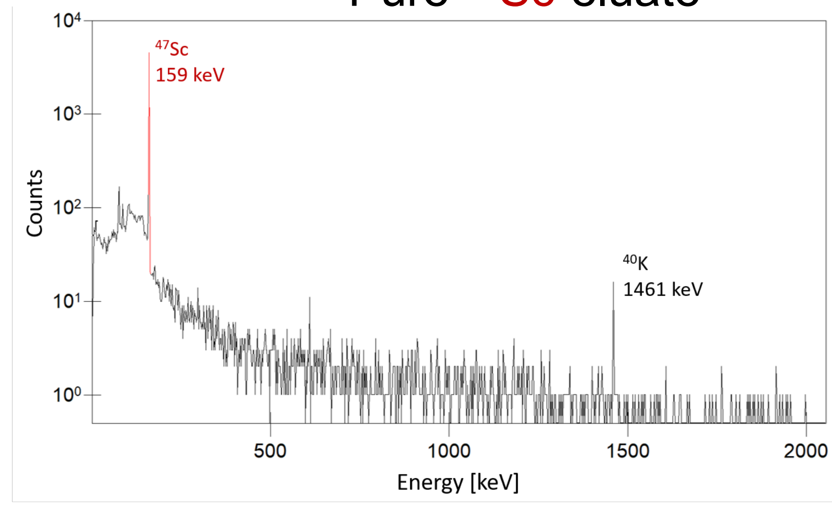


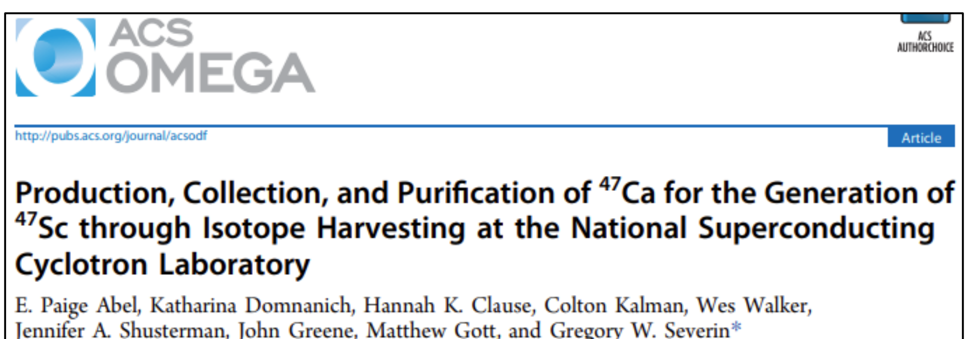
^{47}Ca-fraction





Pure ^{47}Sc-eluate





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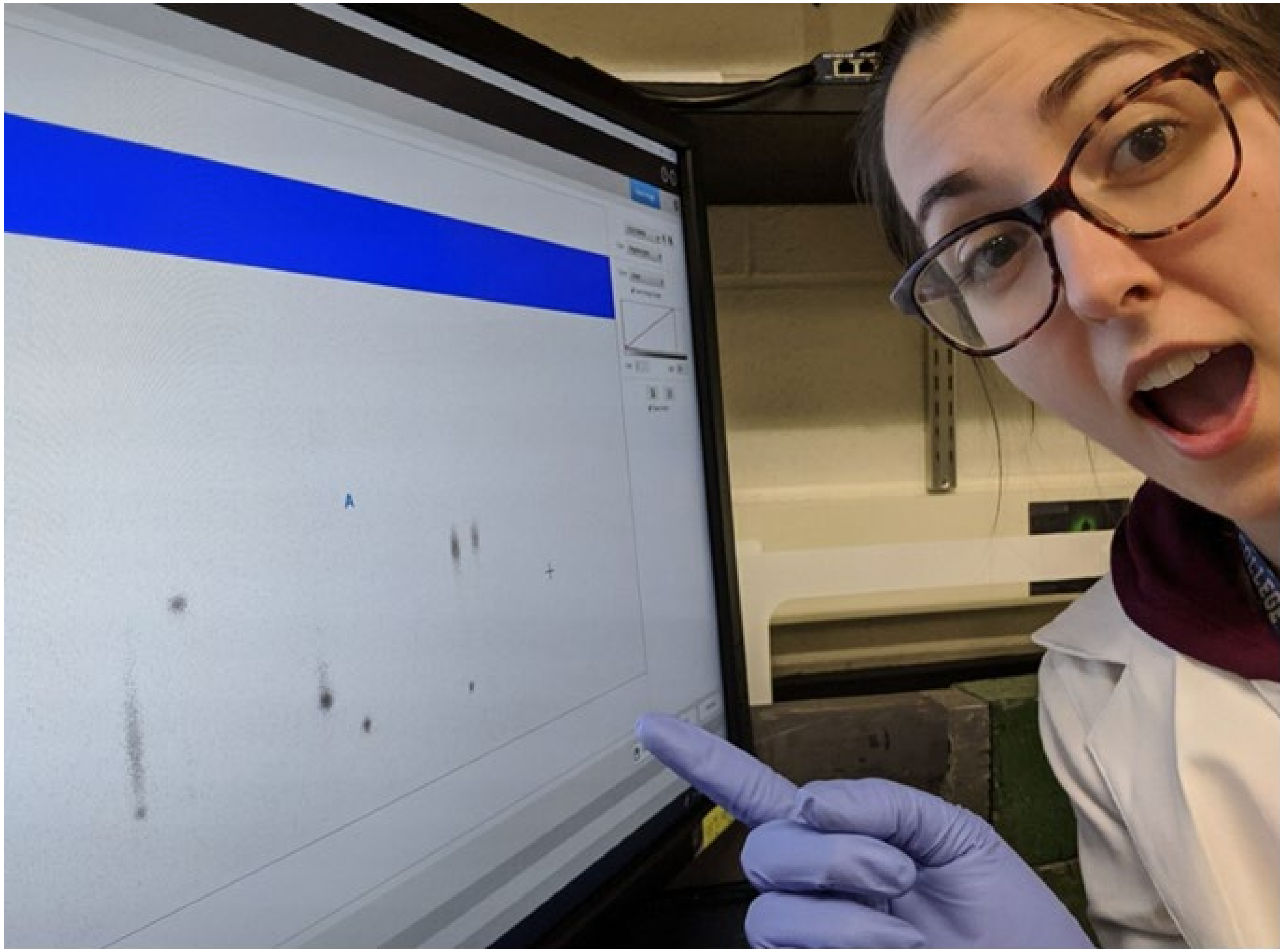
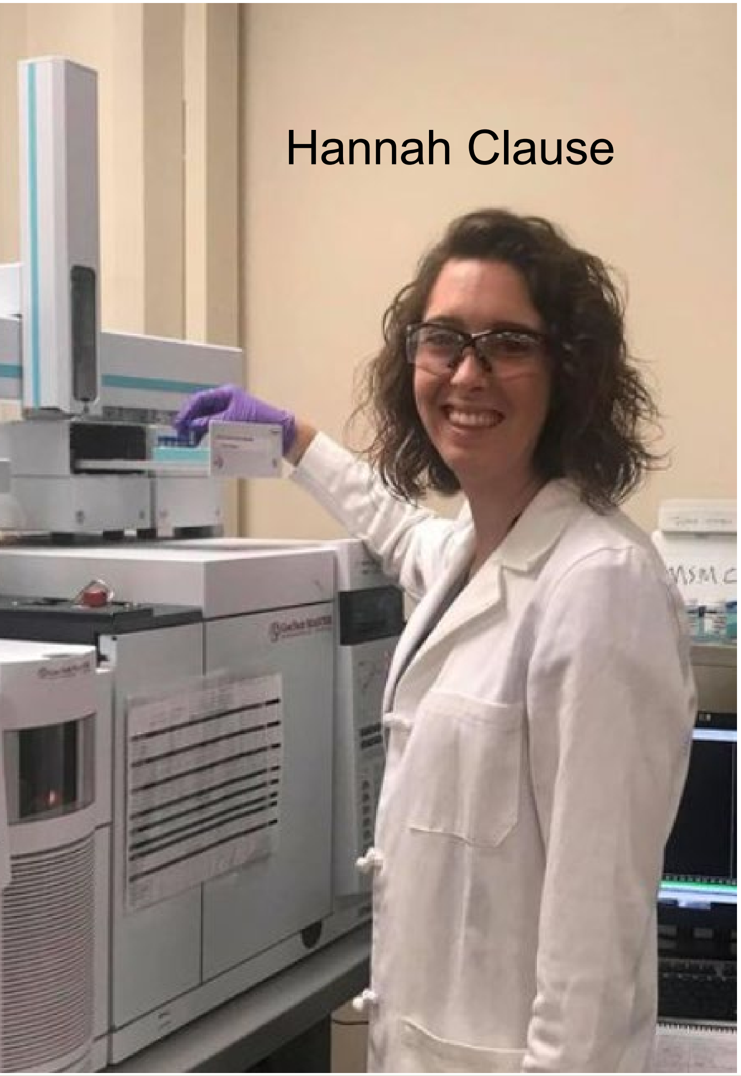
Production, Collection, and Purification of ^{47}Ca for the Generation of ^{47}Sc through Isotope Harvesting at the National Superconducting Cyclotron Laboratory

E. Paige Abel, Katharina Domnanich, Hannah K. Clause, Colton Kalman, Wes Walker, Jennifer A. Shusterman, John Greene, Matthew Gott, and Gregory W. Severin*

Calcium-48 Beam – Radiolabeling
^{47}Sc-DTPA-TOC

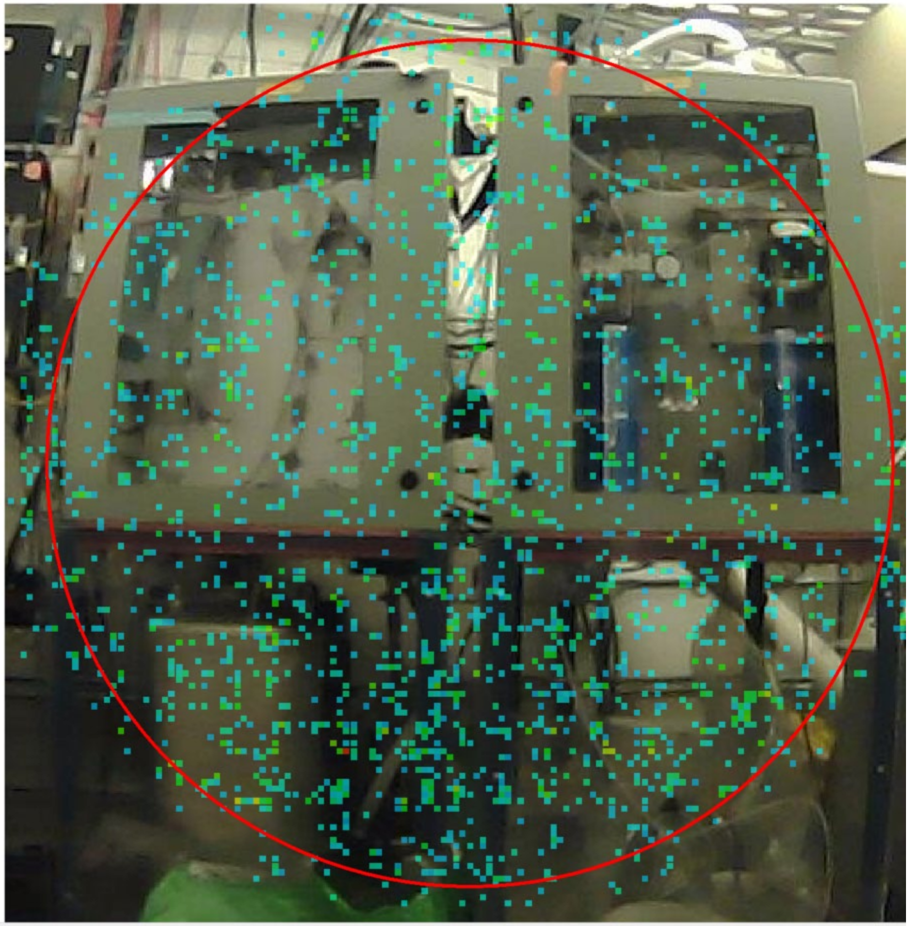


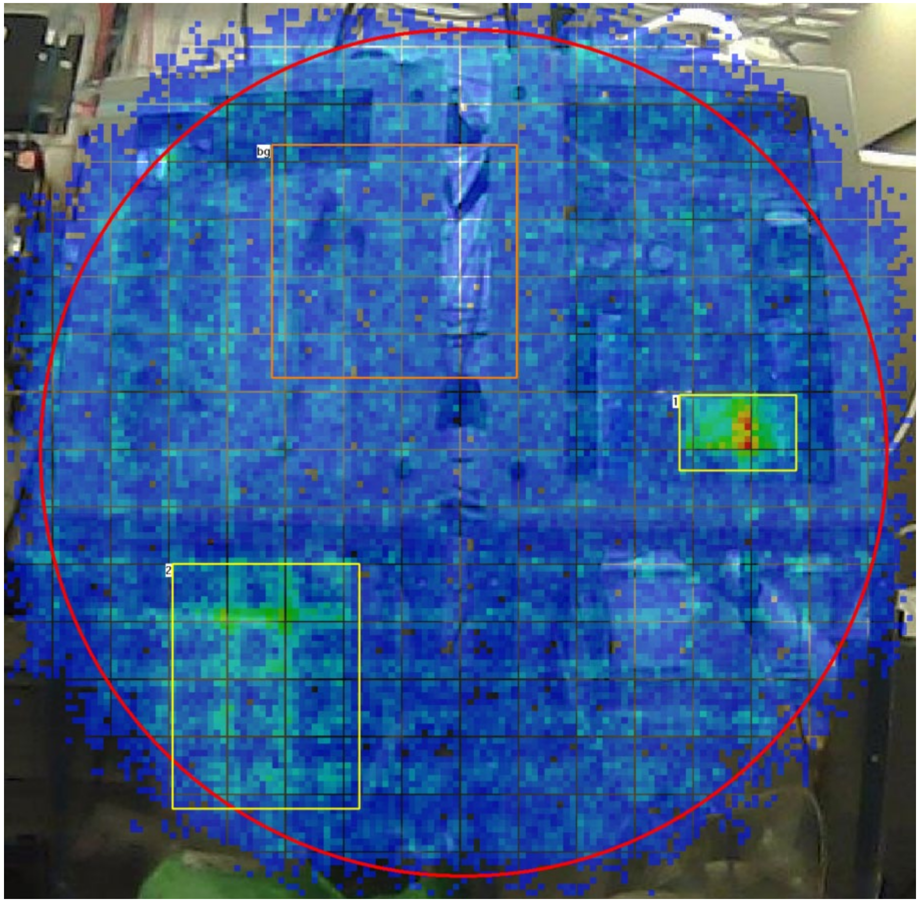
Krypton-78 Beam



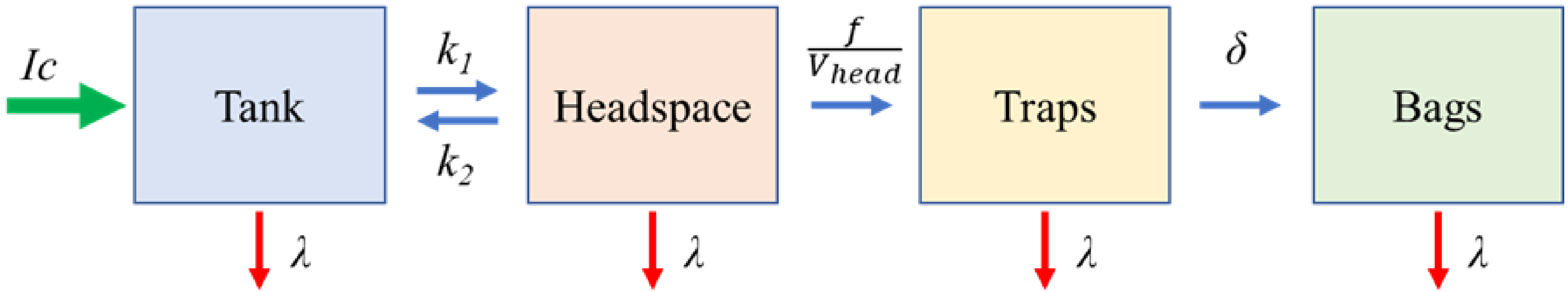
	t1/2		uCi measured	uCi 1:1	#per 78Kr
76Kr	14.8	h	1.02E+02	5.83E+04	1.75E-03
76Br	16.2	h	9.60E+01	5.40E+04	1.78E-03
77Br	57.036	h	4.40E+01	1.68E+04	2.62E-03
73Se	7.15	h	1.20E+02	1.05E+05	1.15E-03
66Ga	9.304	h	5.12E+01	8.57E+04	5.97E-04
72As	26	h	2.19E+01	3.53E+04	6.19E-04
69Ge	39.05	h	1.59E+01	2.41E+04	6.59E-04
71As	65.3	h	1.31E+01	1.47E+04	8.92E-04
75Br	1.611	h	4.73E+02	2.33E+05	2.03E-03
77Kr	74.4	m	1.08E+03	2.50E+05	4.32E-03
79 Kr	35	h	1.50E+01		3.34E-04
7Be	53.22	d	5.81E+00	7.77E+02	7.48E-03
67Ga	3.2617	d	6.09E+00	1.24E+04	4.93E-04
24Na	14.997	h	7.14E+00	5.77E+04	1.24E-04
61Co	1.649	h	1.62E+02	2.32E+05	7.01E-04
62Zn	9.186	h	7.61E+00	8.66E+04	8.78E-05
72Se	8.4	d	2.62E+00	4.87E+03	5.38E-04
52Mn	5.591	d	2.14E+00	7.28E+03	2.94E-04
61Cu	3.339	h	1.77E+01	1.72E+05	1.03E-04
81Rb	4.572	h	1.01E+01	1.43E+05	7.09E-05
83Sr	32.41	h	2.03E+00	2.88E+04	7.06E-05
56Mn	2.5789	h	8.64E+00	1.94E+05	4.45E-05
80Sr	106.3	m	6.33E+00	2.26E+05	2.80E-05
47Sc	3.3492	d	5.00E-02	1.20E+04	4.17E-06
70As	52.6	m	1.83E+01	2.69E+05	6.82E-05
74mBr	46	m	3.45E+01	2.76E+05	1.25E-04
63Zn	38.47	m	2.19E+01	2.84E+05	7.71E-05
74Br	25.4	m	3.49E+01	2.97E+05	1.18E-04

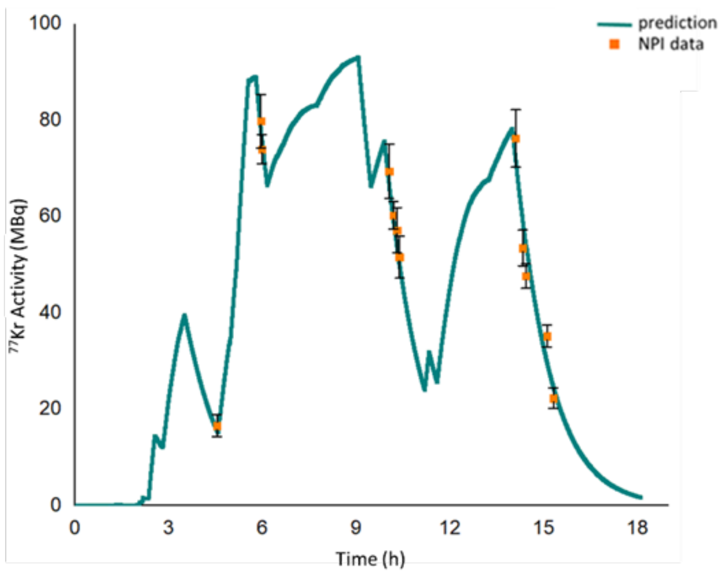
Krypton-78 Beam – isolating ^{radio}Kr



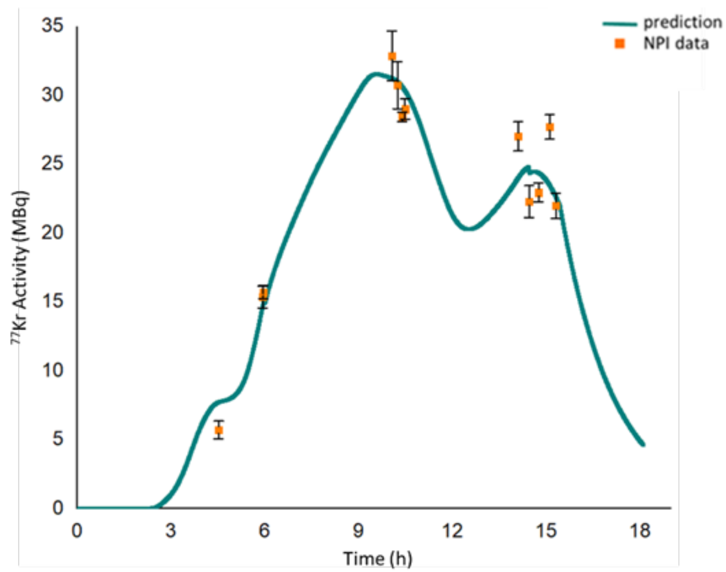


^{77}Kr Mass Transport Results





Tank and
Headspace

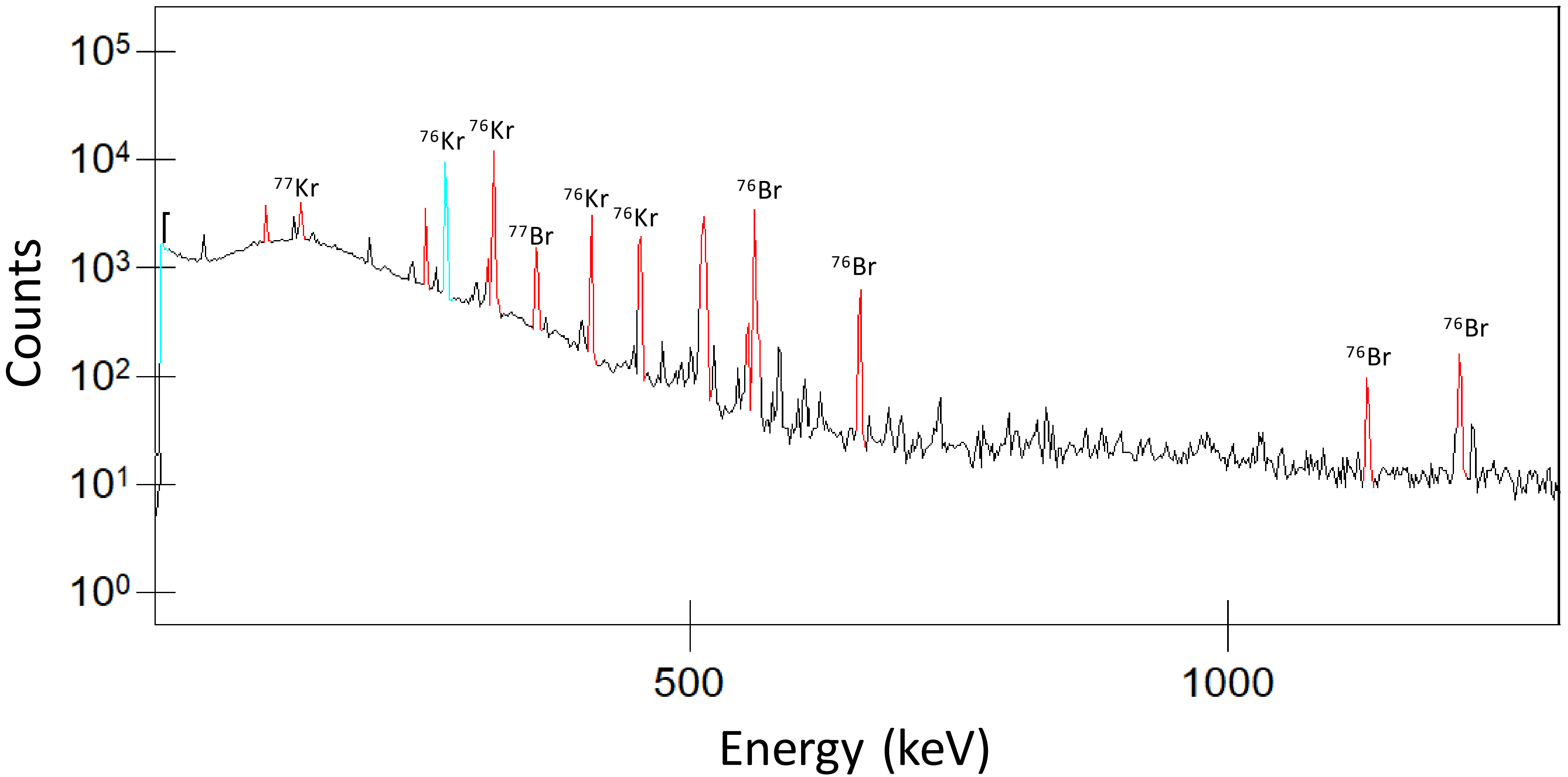


Traps

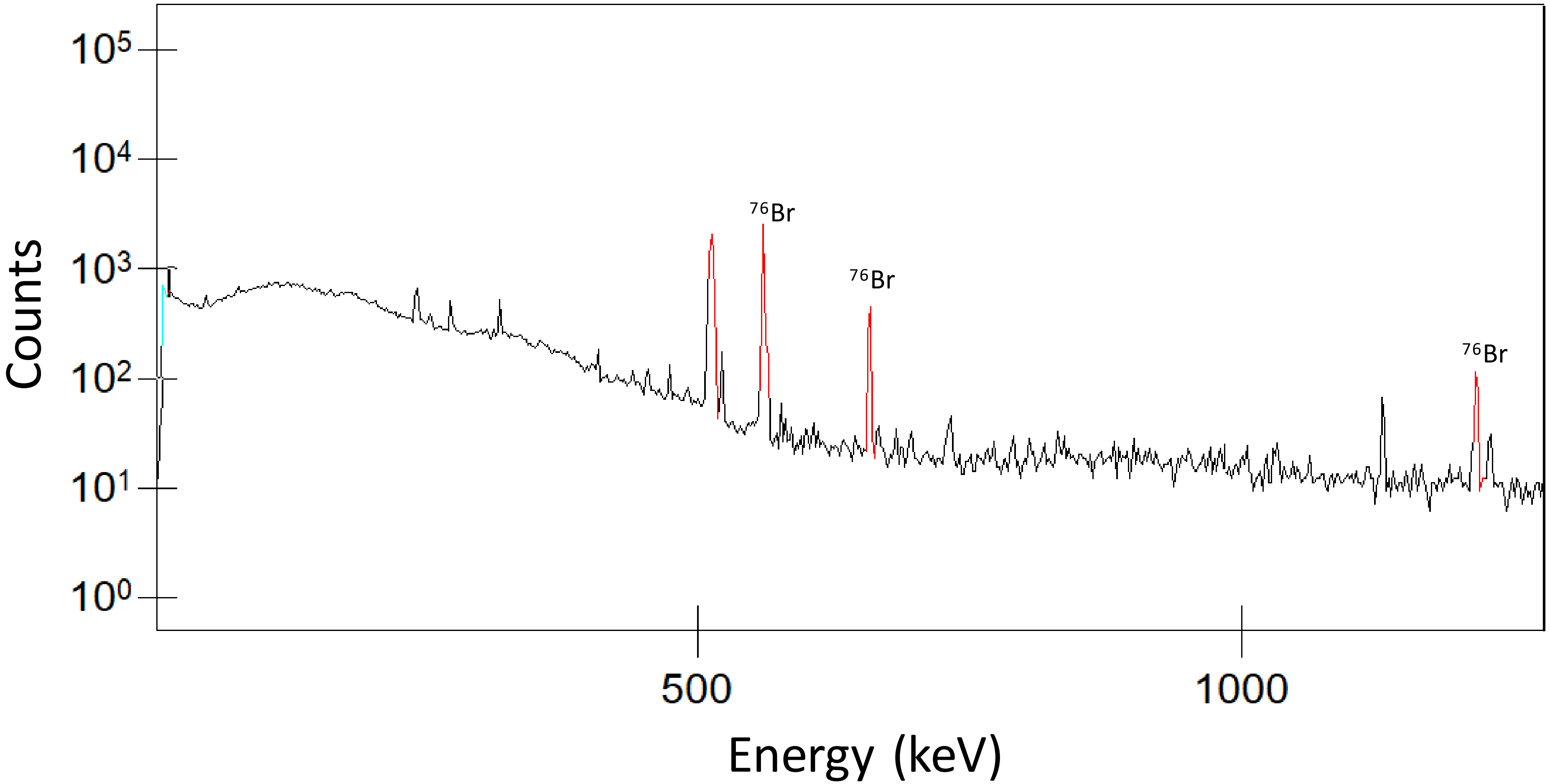
1.18(6) x 10^{-2} nuclei of ^{77}Kr per incident ^{78}Kr ion

Greg Severin, INTDS 202227

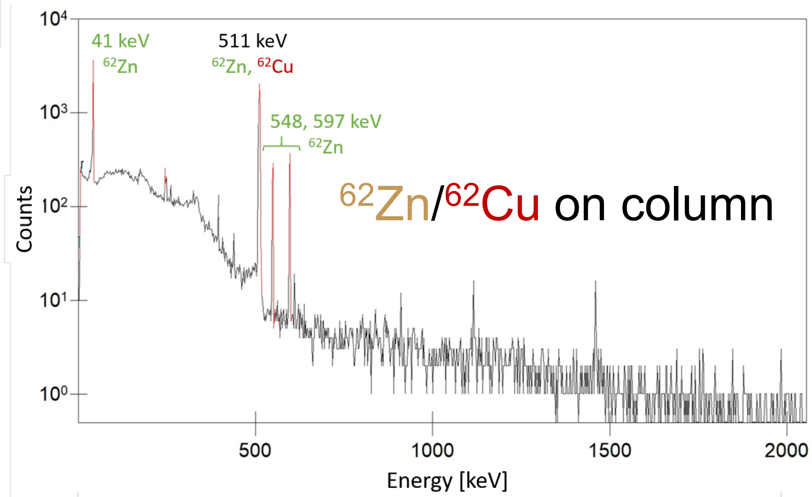
Krypton-78 Beam – Generating pure ^{76}Br

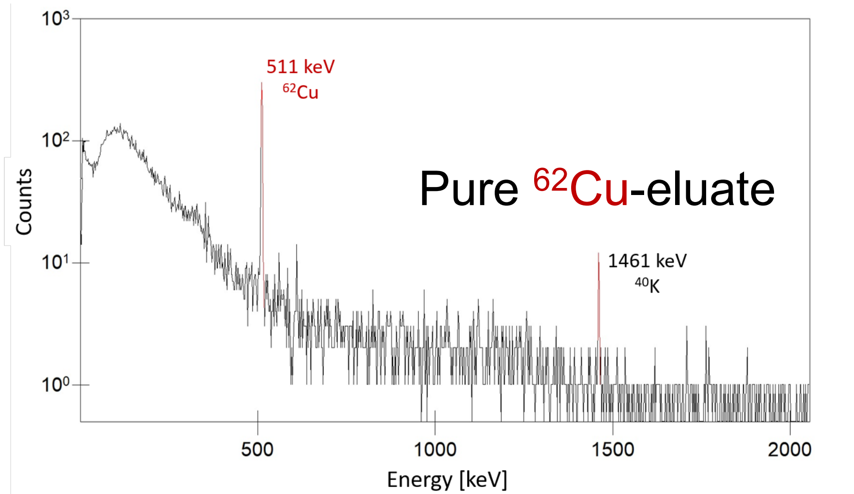


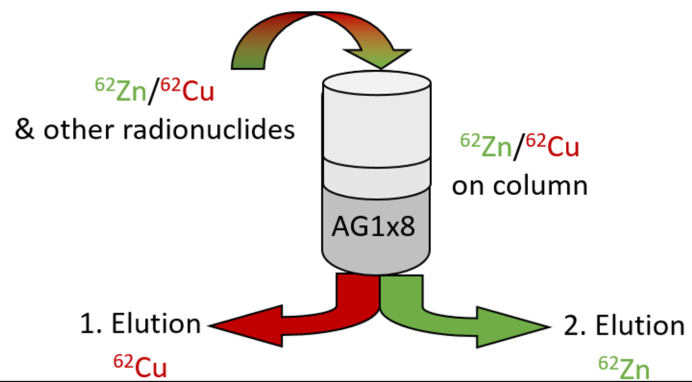
Krypton-78 Beam – Generating pure ^{76}Br



2 in one- ^{62}Cu generator







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Harvesting ^{62}Zn from an aqueous cocktail at the NSCL†

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Conclusions

- Water is an interesting target for heavy ion irradiation
  - We have learned a lot in the 1-500W regime
    - » Radiolysis and Radiation Accelerated Corrosion must be considered
    - » Combining cooling and transport is effective for water soluble products
    - » Some products will be purity limited, but there are several interesting cases where high purity can be reached.
  - Extrapolations look promising for future irradiations at 1000x power, but….
  - We have a lot to learn!

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U.S. Department of Energy Office of Science
Michigan State University

NNSA National Nuclear Security Administration