CYCLOTRON INSTITUTE

Enhanced production of ^{99}Mo in inverse kinematics heavy ion reactions

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HINPw6 – 14-16 May 2021

6th Workshop of the Hellenic Institute of Nuclear Physics

OUTILNE

- Introduction
- Experiment
- Data Analysis
- Results and Discussion
- Summary

INTRODUCTION

Methodology → **Radioisotope production using inverse kinematics**

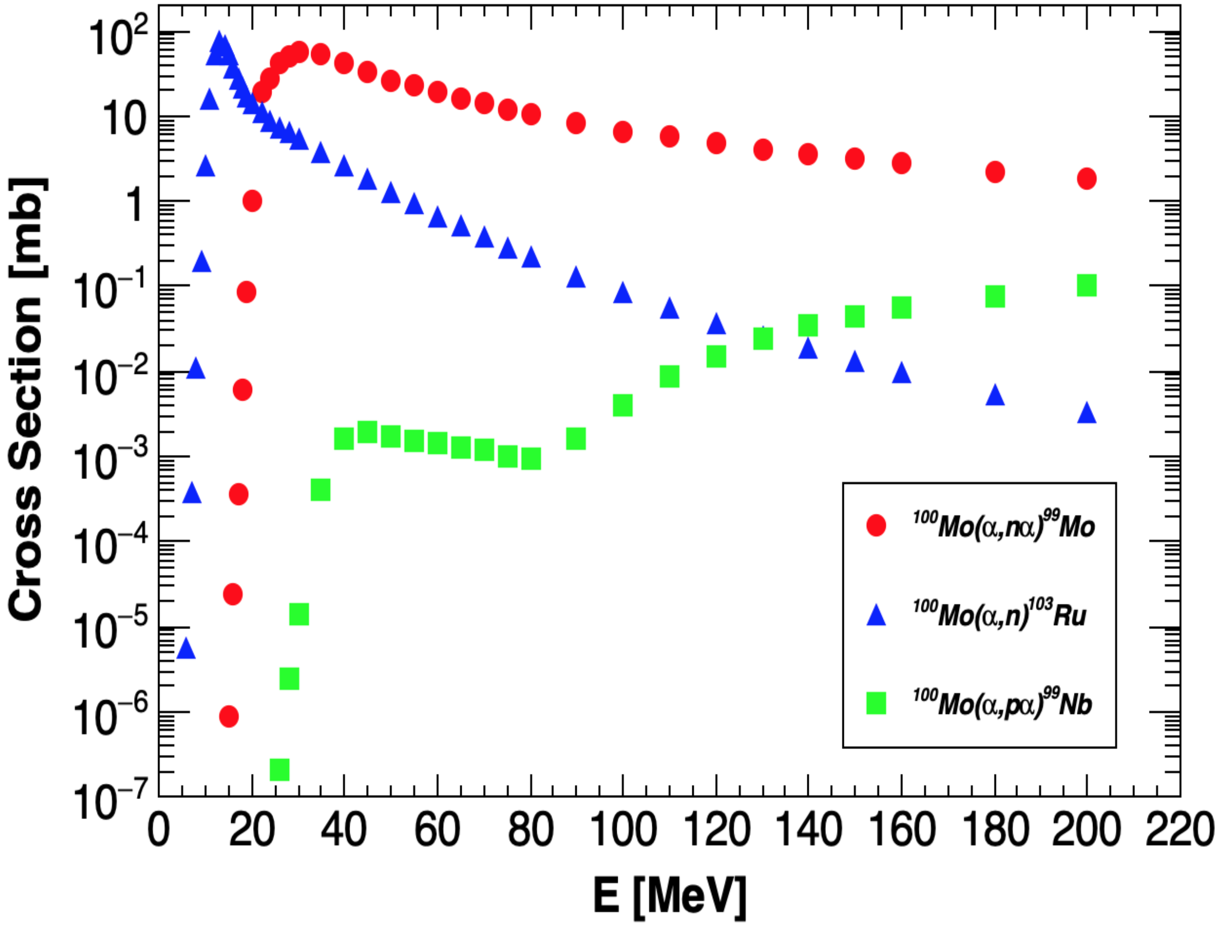
- Successful test of production of the theranostic radionuclide ^{67}Cu ($T_{1/2}$ = 62 h)
- Reaction of a ^{70}Zn beam at 15 MeV/A with a hydrogen gas target
- ^{67}Cu alongside other coproduced isotopes, collected after the gas target on an Al catcher foil and radioactivity measured by off-line γ-ray analysis.
- Use of secondary emitted particles from the primary nuclear reaction to irradiate other targets

INTRODUCTION

- Investigation of alternative production method for important isotopes for nuclear medicine in high demand worldwide
- Production in reactors not enough to supply the demand
- ^{99}Mo/^{99m}Tc generator: world demand estimated to ~ 7 kCi/week
- ^{99m}Tc: 140 keV γ-ray emitter (I_γ = 89%, $T_{1/2}$ = 6.01 h) considered to be ideal radiotracer and used in ~85% of all nuclear medicine diagnostic scans worldwide
- ^{99m}Tc is produced via β-decay from ^{99}Mo ($T_{1/2}$ = 65.94 h)
- Other accelerator-made of ^{99}Mo/^{99m}Tc : ^{100}Mo(p,2n)^{99m}Tc and ^{100}Mo(p,pn)^{99}Mo

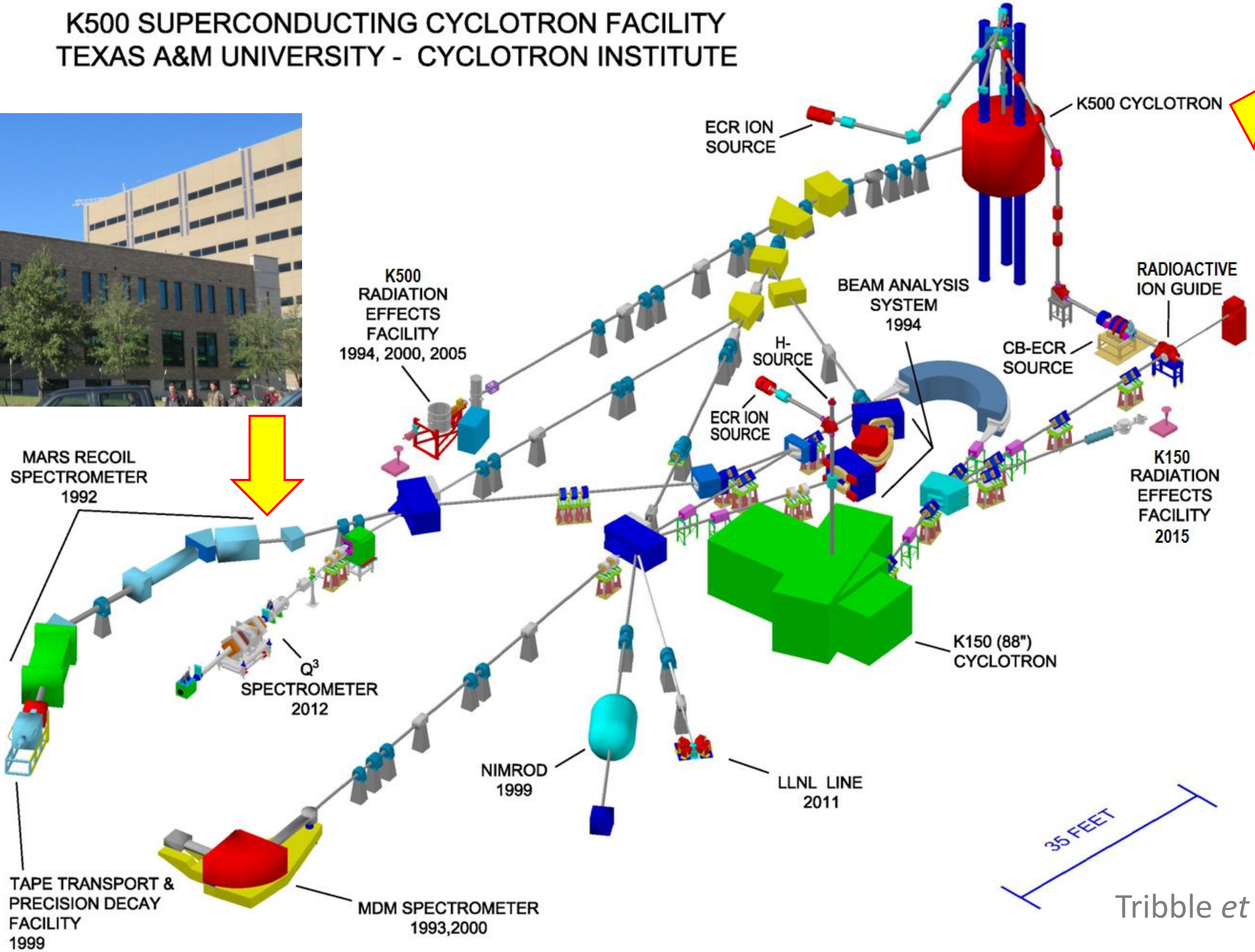
INTRODUCTION

- Cross-section estimates from **TALYS** for different channels.
- Nucleon transfer on a ^{4}He gas target produces resonances in ^{5}He (^{5}Li), and possibly high cross-section
- ^{100}Mo+^{4}He→^{5}He (^{5}Li)+^{99}Mo (^{99}Nb) Q=-9.2MeV (-13.1 MeV)



https://tendl.web.psi.ch/tendl_2019/tendl2019.html

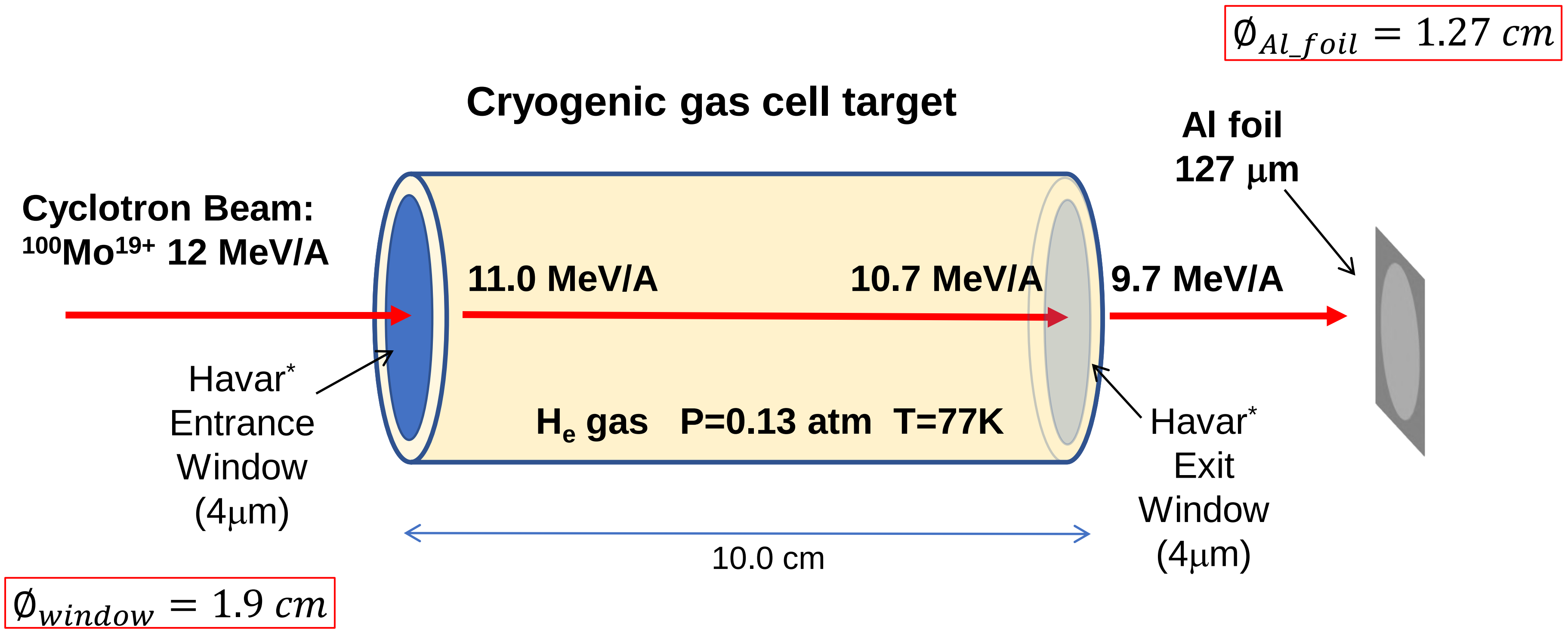
EXPERIMENT



12 MeV/A beam of $^{100}Mo_{19+}$

Tribble *et al.*, NIMA 285 (1989) 441-446

Experimental Setup



$\emptyset_{Al\_foil} = 1.27\ cm$

Cryogenic gas cell target

Al foil
127 μm

Cyclotron Beam:
$^{100}Mo^{19+}$ 12 MeV/A

11.0 MeV/A

10.7 MeV/A

9.7 MeV/A

Havar*
Entrance
Window
(4μm)

H_e gas P=0.13 atm T=77K

Havar*
Exit
Window
(4μm)

10.0 cm

$\emptyset_{window} = 1.9\ cm$

*HAVAR® - High strength non-magnetic alloy foil
Co 42.0% Mo 2.2% Cr 19.5% Mg 1.6% Ni 12.7% C 0.2% W 2.7% Fe Balance

Souliotis *et al.,* Appl. Radiat. Isot. 149 (2019) 89-95

Experimental Specifications

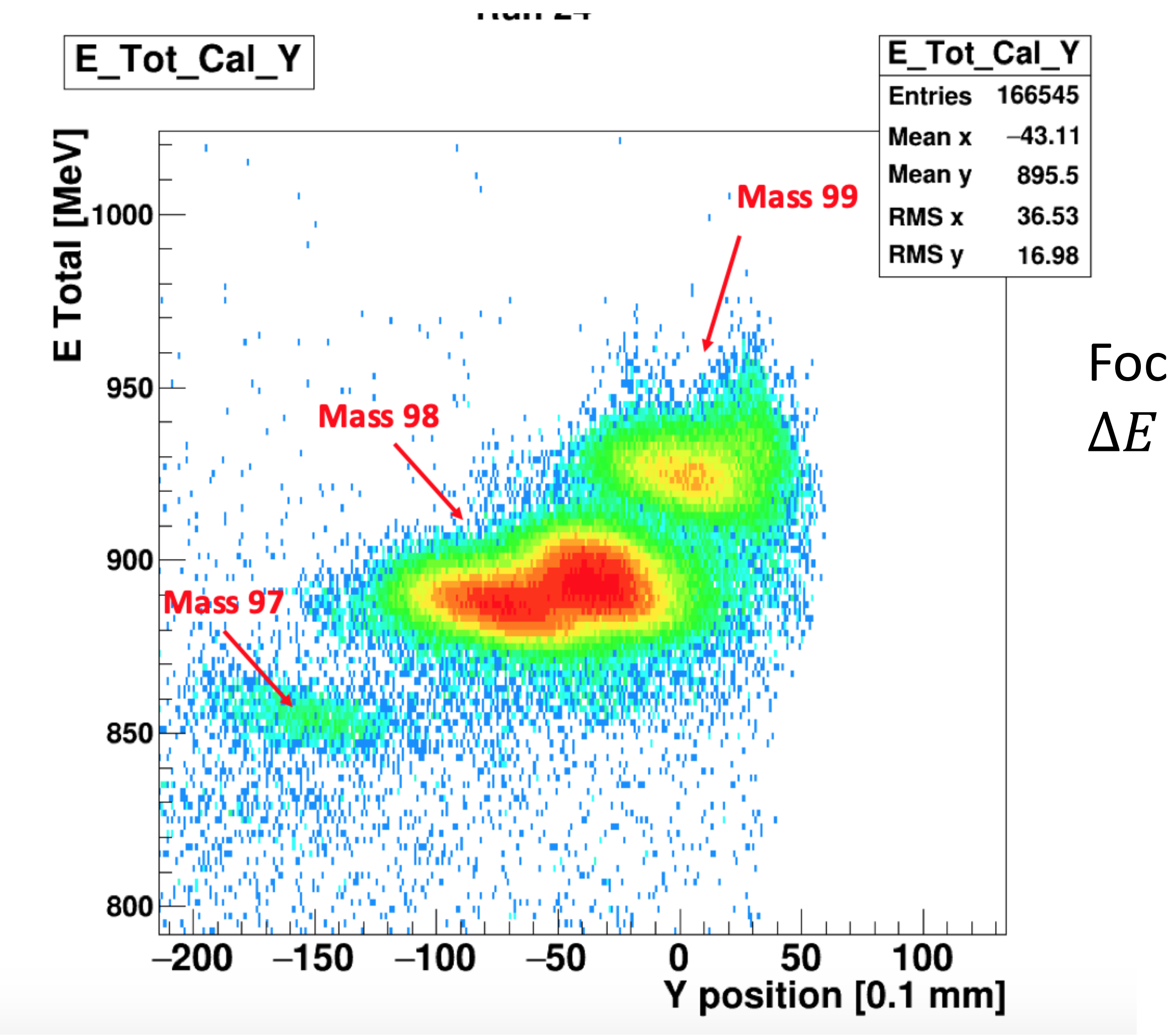
Irradiation

- ^{100}Mo (12 MeV/A) at an average charge state of 35$^+$
- 3 different runs
  - Source 1 (102 torr, 0.07 pnA, 11h35)
  - Source 2 (213 torr, 0.21pnA, 10h28);
  - Source 3 (1008 torr, 0.17 pnA, 7h52)
- Beam current was periodically monitored and was nearly constant (within 15 %)

Off-line γ-ray analysis

- HPGe detector
- Al foil @ d = 50 mm
- dead time 2-3%
- Energy resolution 2.5–4.0 keV (FWHM)

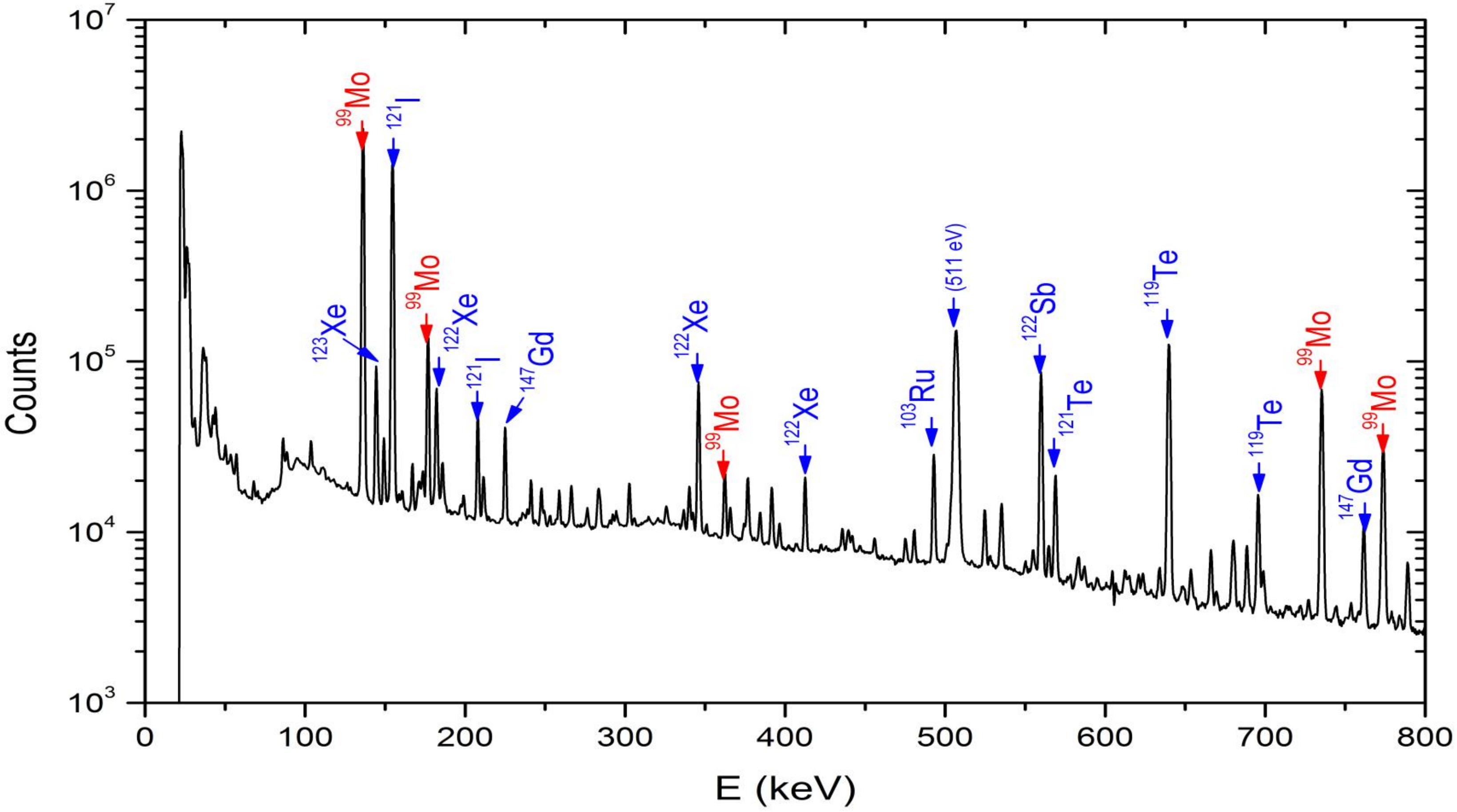
DATA ANALYSIS



Focal plane of MARS

$\Delta E - E$, Si detector 55 μm-500 μm

γ −spectra from from source 3



Activity after irradiation

Averaged Observed Activity

$$R_M = \frac{C}{\varepsilon \Delta t_L I_\gamma}$$

Net area of the peak

Efficiency

Measurement time

Branching Ratio

Activity after Irradiation

Cool time

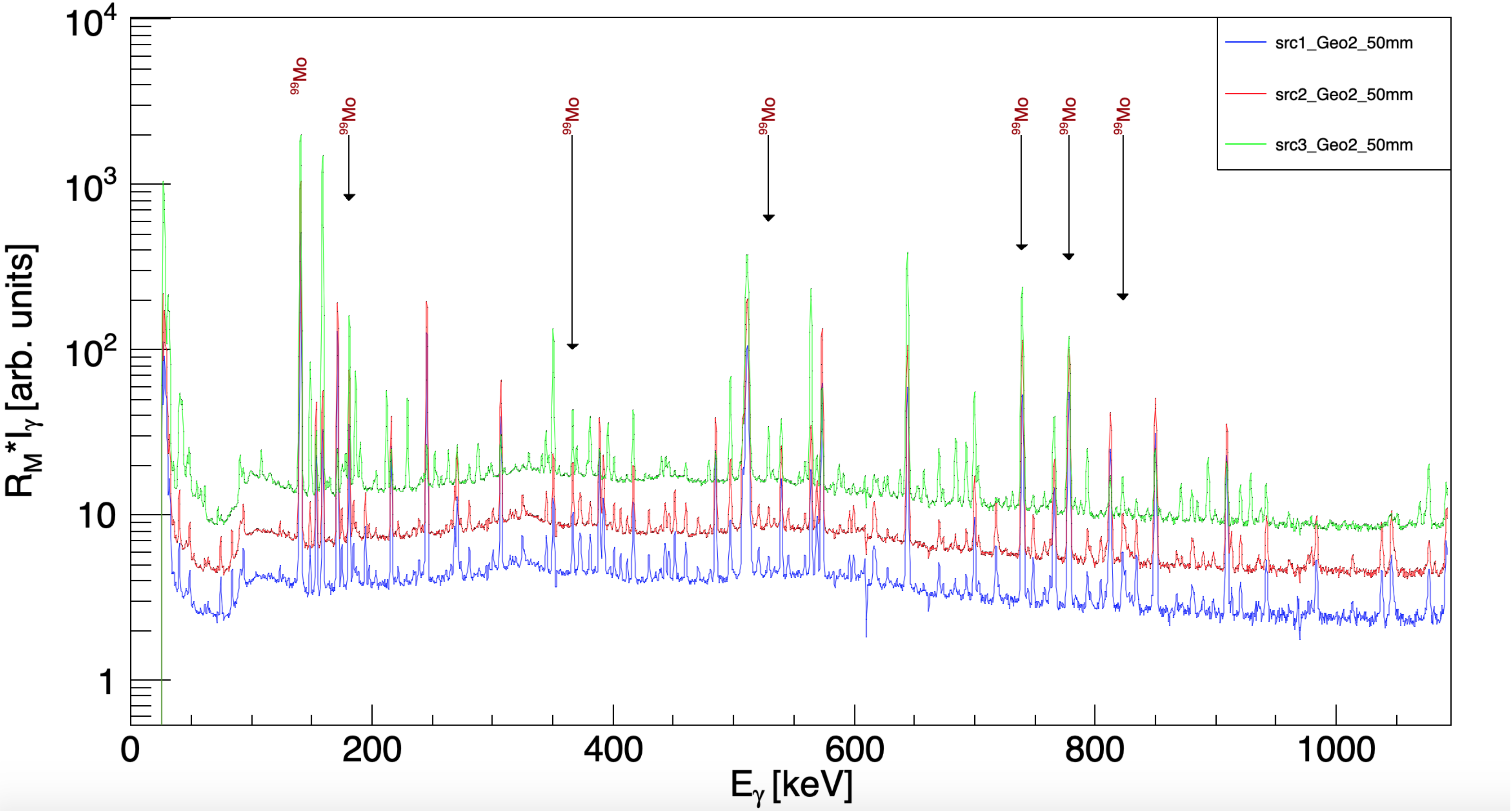
Real time

Decay Constant

Beam time

$$R_{AI} = R_M \underbrace{e^{\lambda t_{dk}}}_{\text{Correction for activity drop during Cool time}} \underbrace{\frac{\lambda \Delta t_R}{\left(1 - e^{-\lambda \Delta t_R}\right)}}_{\text{Correction for decay during measurement}} \underbrace{\frac{\lambda t_{irr}}{\left(1 - e^{-\lambda t_{irr}}\right)}}_{\text{Correction for decay during irradiation}}$$

γ −spectra from 3 runs with different He gas densities



Activity after irradiation

$$R_{AI} = R_M \, e^{\lambda t_{dk}} \frac{\lambda \Delta t_R}{\left(1 - e^{-\lambda \Delta t_R}\right)} \frac{\lambda t_{irr}}{\left(1 - e^{-\lambda t_{irr}}\right)}$$

Net Activity = Activity produced per beam intensity unit and irradiation time

$$H_{AI} = \frac{R_{AI}}{I_{beam} t_{irr}}$$

Activities for different isotopes of interest

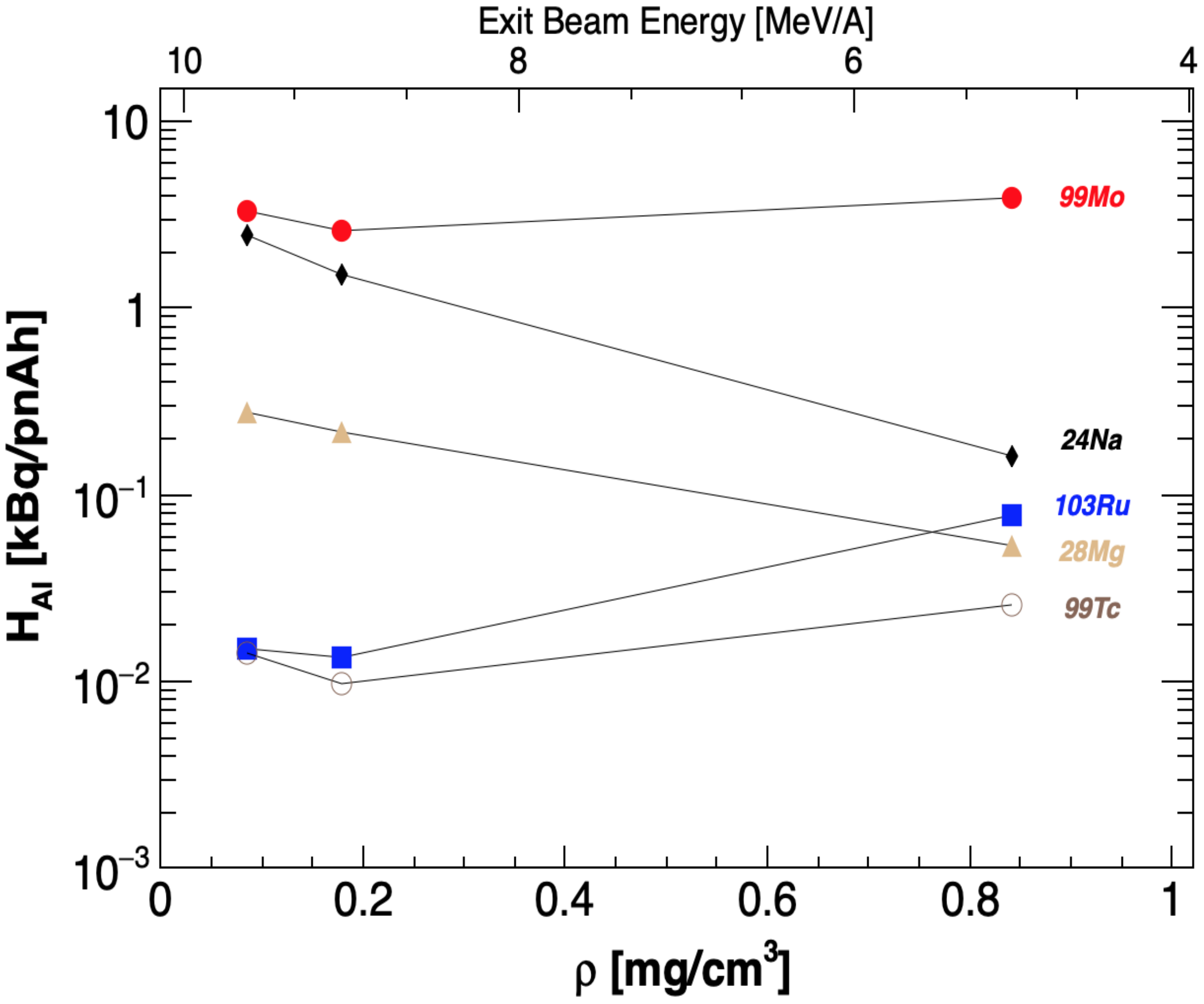
- Identified using their strongest and independent γ-lines

$^{100}Mo+^{4}He \rightarrow {}^{5}He\ ({}^{5}Li)+{}^{99}Mo\ ({}^{99}Nb)$
Q=-9.2 MeV (-13.1 MeV)

$^{100}Mo+^{27}Al \rightarrow {}^{28}Al({}^{28}Si,{}^{28}Mg)+{}^{99}Mo({}^{99}Nb,{}^{99}Tc)$
Q=-0.57 MeV(0.4MeV,-1.0 MeV)

$^{100}Mo+^{4}He \rightarrow {}^{24}Na+{}^{80}As$ (Q=-3.1 MeV)
$^{100}Mo+^{27}Al \rightarrow {}^{24}Na+{}^{103}Ru$ (Q=-7.7 MeV)

$^{100}Mo+^{4}He \rightarrow n+{}^{103}Ru$ Q=-4.6 MeV



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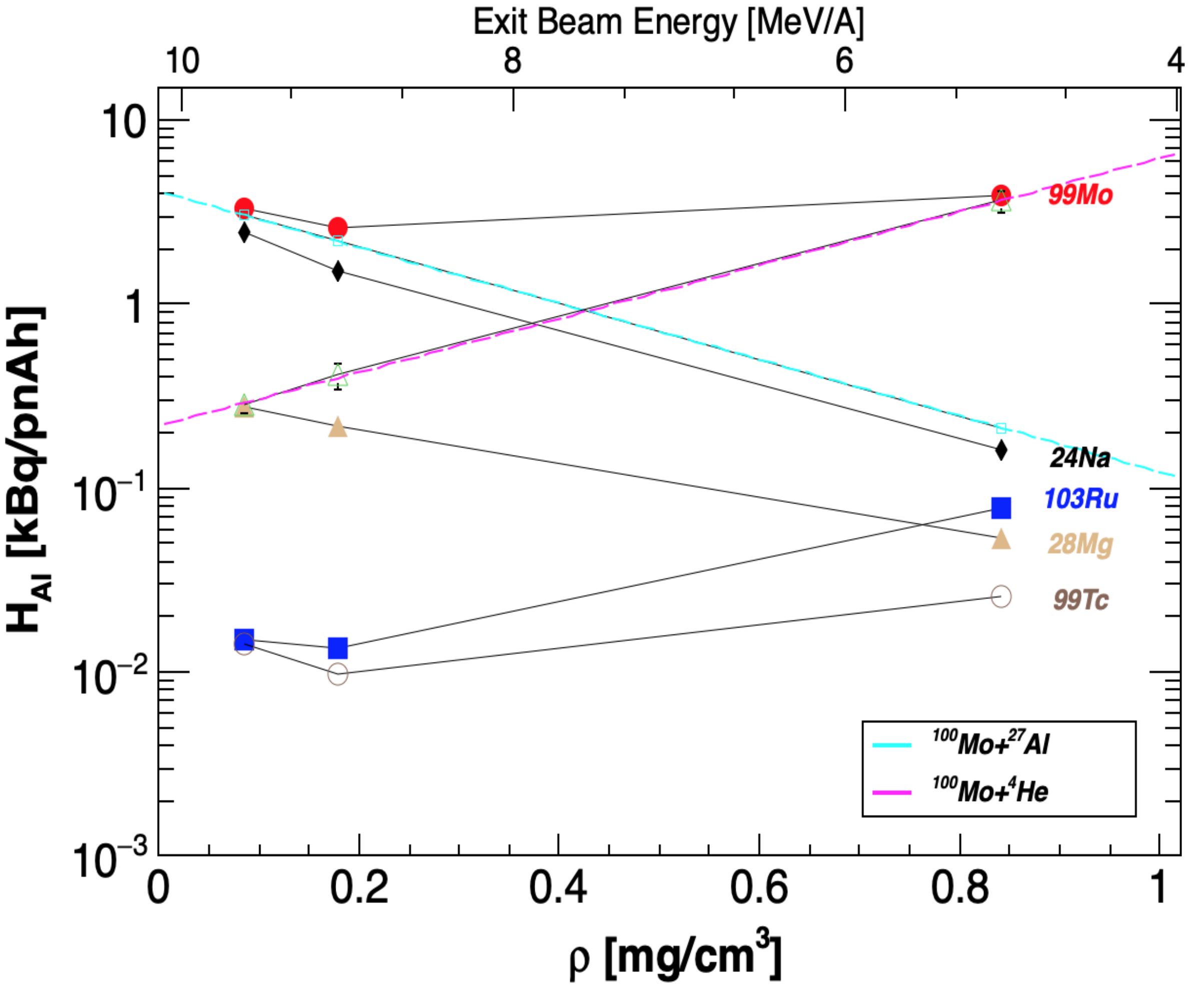

SUMMARY

- This work has demonstrated the feasibility and advantages of using inverse kinematics to produce ^{99}Mo and other isotopes.
- A gas target and a catcher have been used for production.
- Neutrons produced in the forward direction could be used to irradiate other targets and produce a variety of radioisotopes of interest.
- Production could also be determined for neutrons in coincidence with heavy fragments detected in MARS.
- Currently working on the extraction of cross-sections to compare to data if available and to theoretical models.

Collaboration

Cyclotron Institute, Texas A&M University, USA

Dr. Aldo Bonasera
Dr. Marcia R. D. Rodrigues
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University of Athens, Greece

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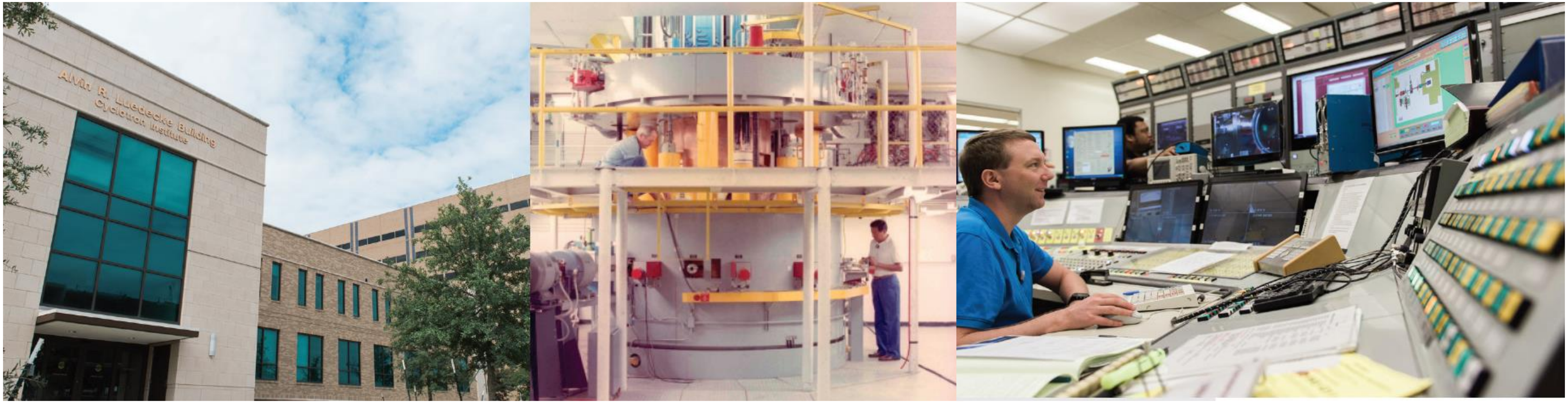
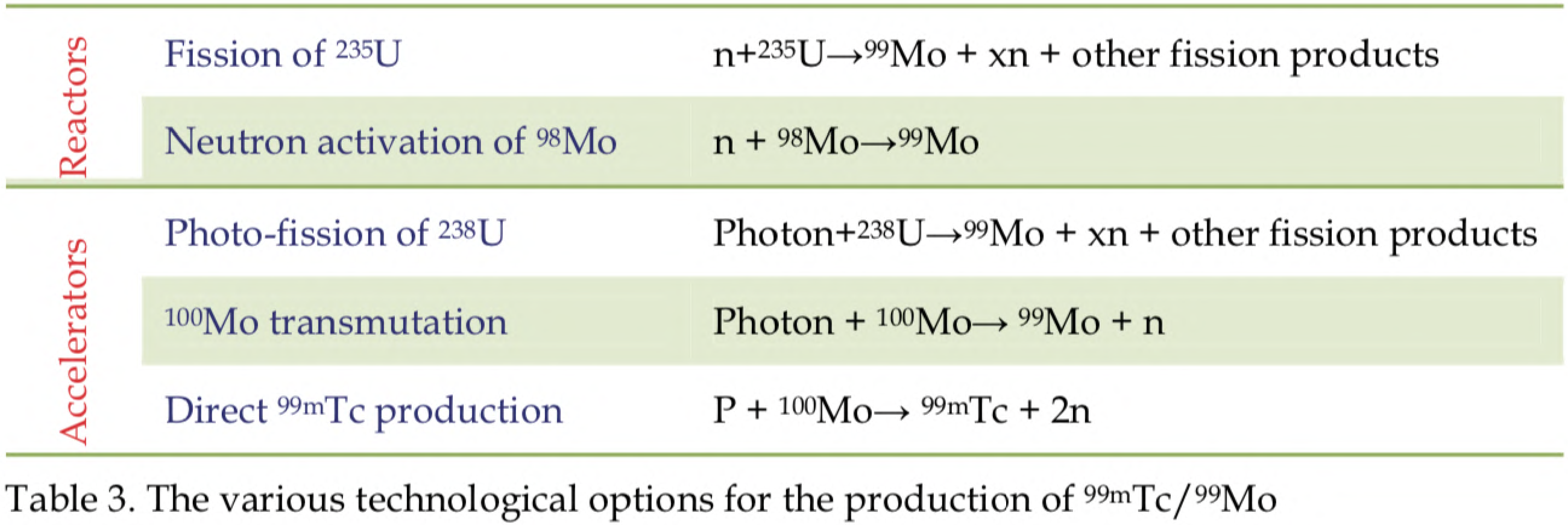


Thank you for your attention

Back up slides

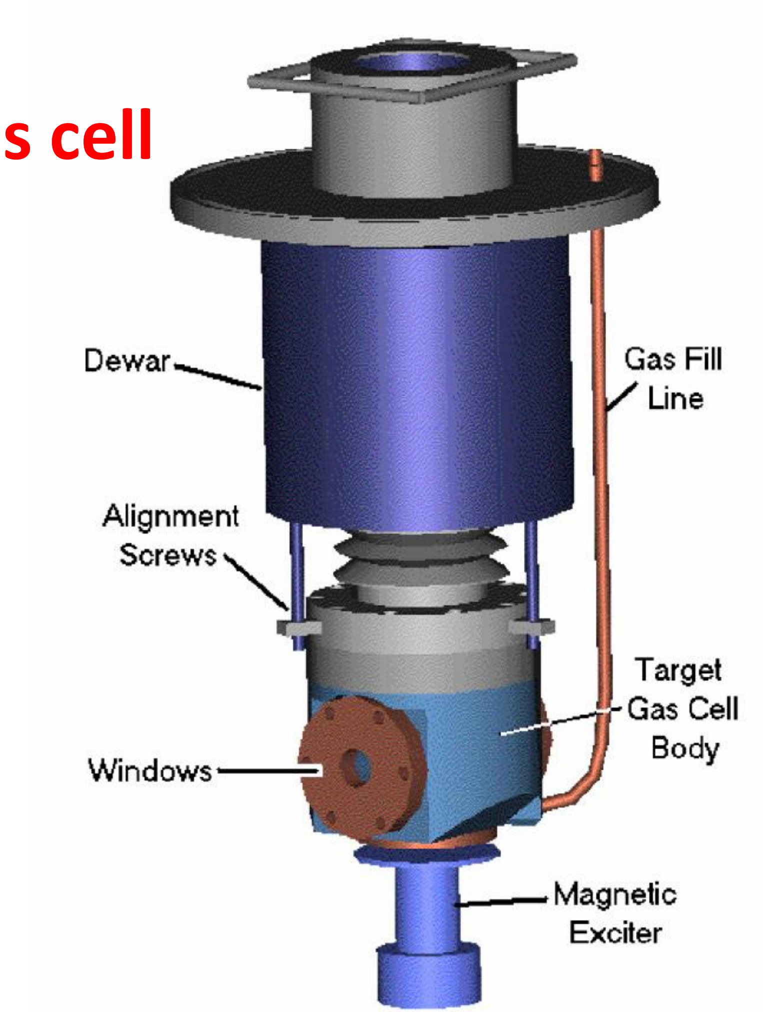


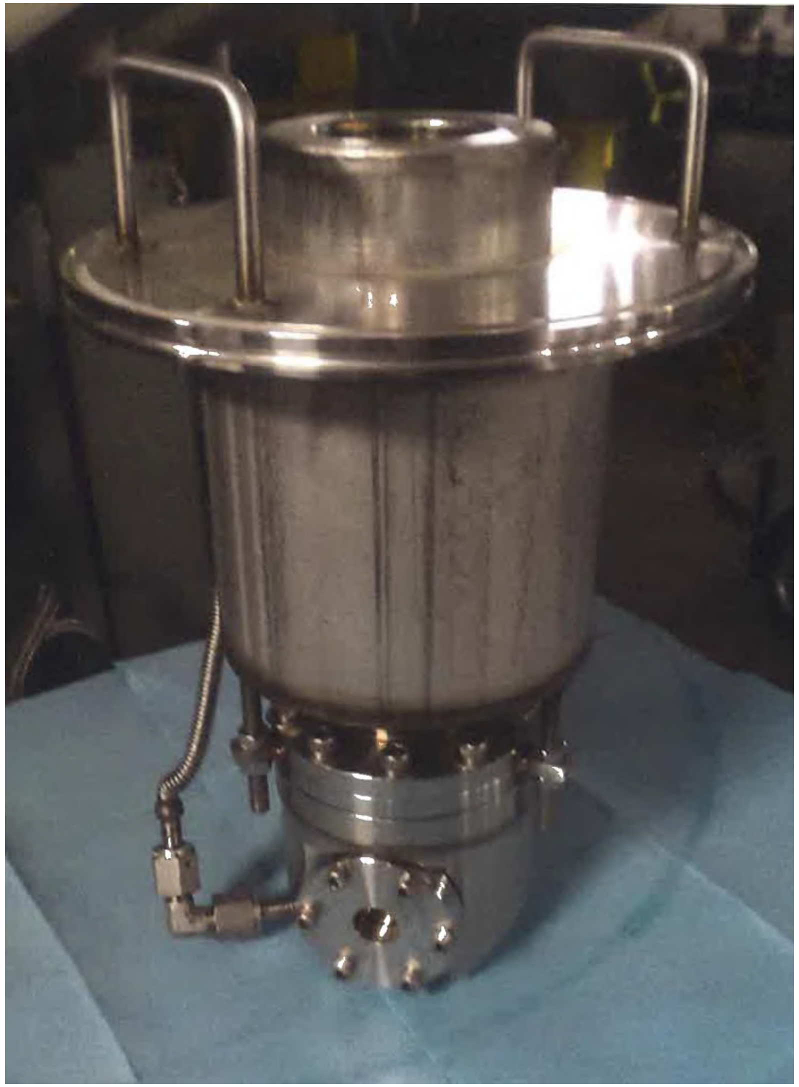
Reactors	Fission of ^{235}U	$n+{}^{235}\mathrm{U}\rightarrow{}^{99}\mathrm{Mo} + xn$ + other fission products
	Neutron activation of ^{98}Mo	$n + {}^{98}\mathrm{Mo}\rightarrow{}^{99}\mathrm{Mo}$
Accelerators	Photo-fission of ^{238}U	Photon$+{}^{238}\mathrm{U}\rightarrow{}^{99}\mathrm{Mo} + xn$ + other fission products
	^{100}Mo transmutation	Photon $+ {}^{100}\mathrm{Mo}\rightarrow {}^{99}\mathrm{Mo} + n$
	Direct ^{99m}Tc production	$P + {}^{100}\mathrm{Mo}\rightarrow {}^{99m}\mathrm{Tc} + 2n$

Table 3. The various technological options for the production of ^{99m}Tc/^{99}Mo

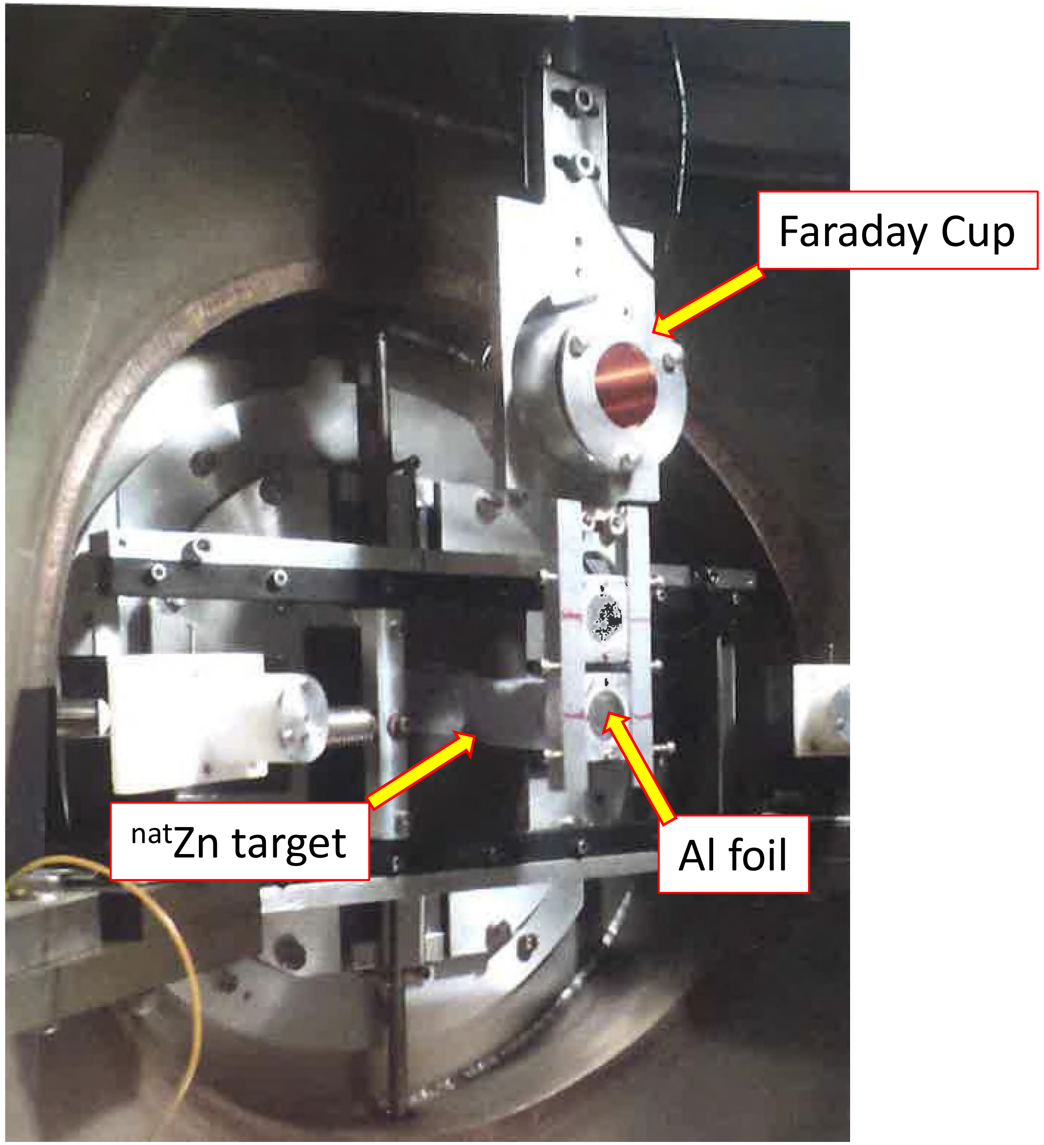
Experimental Set up

Cryogenic gas cell

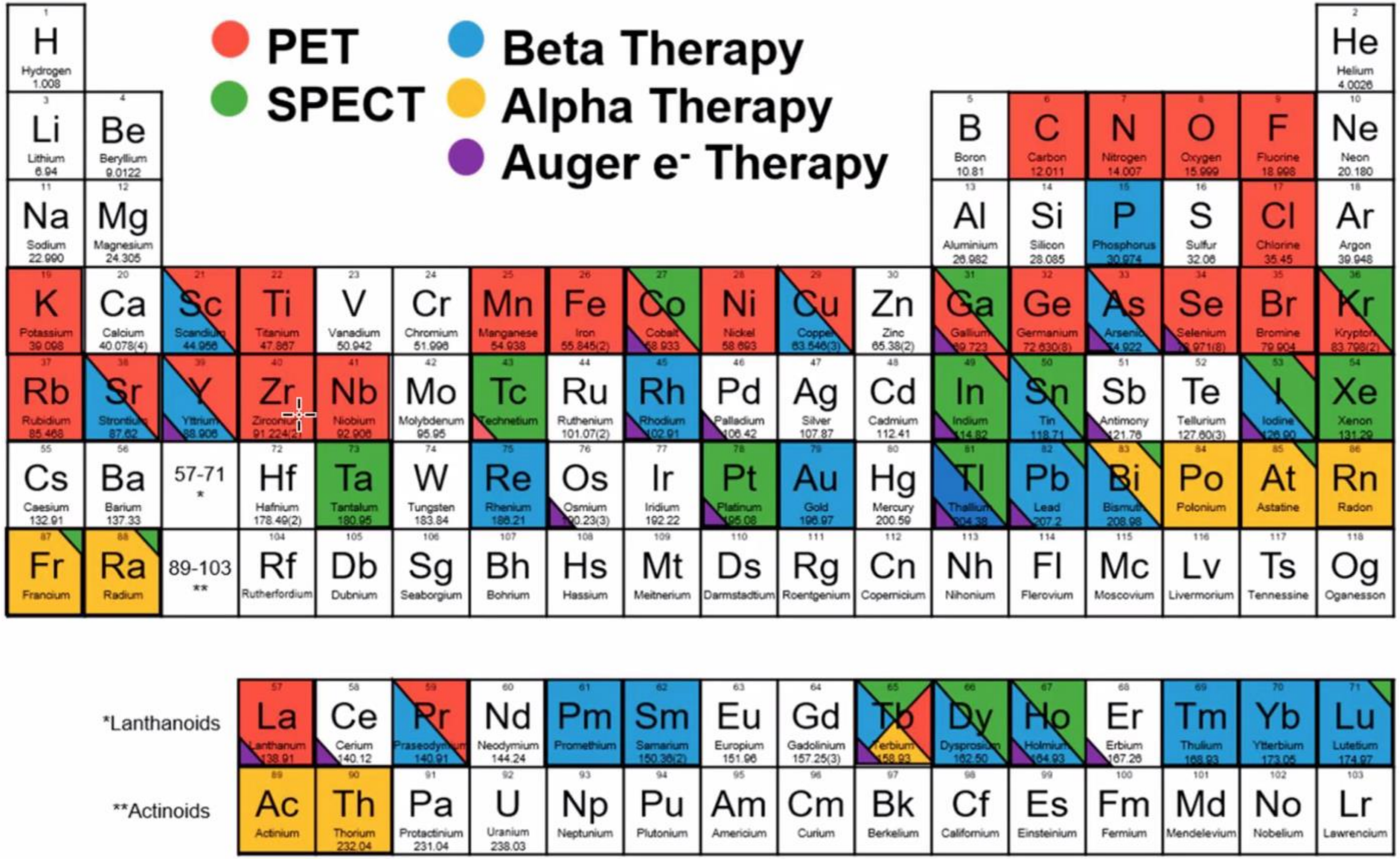




Experimental Set up

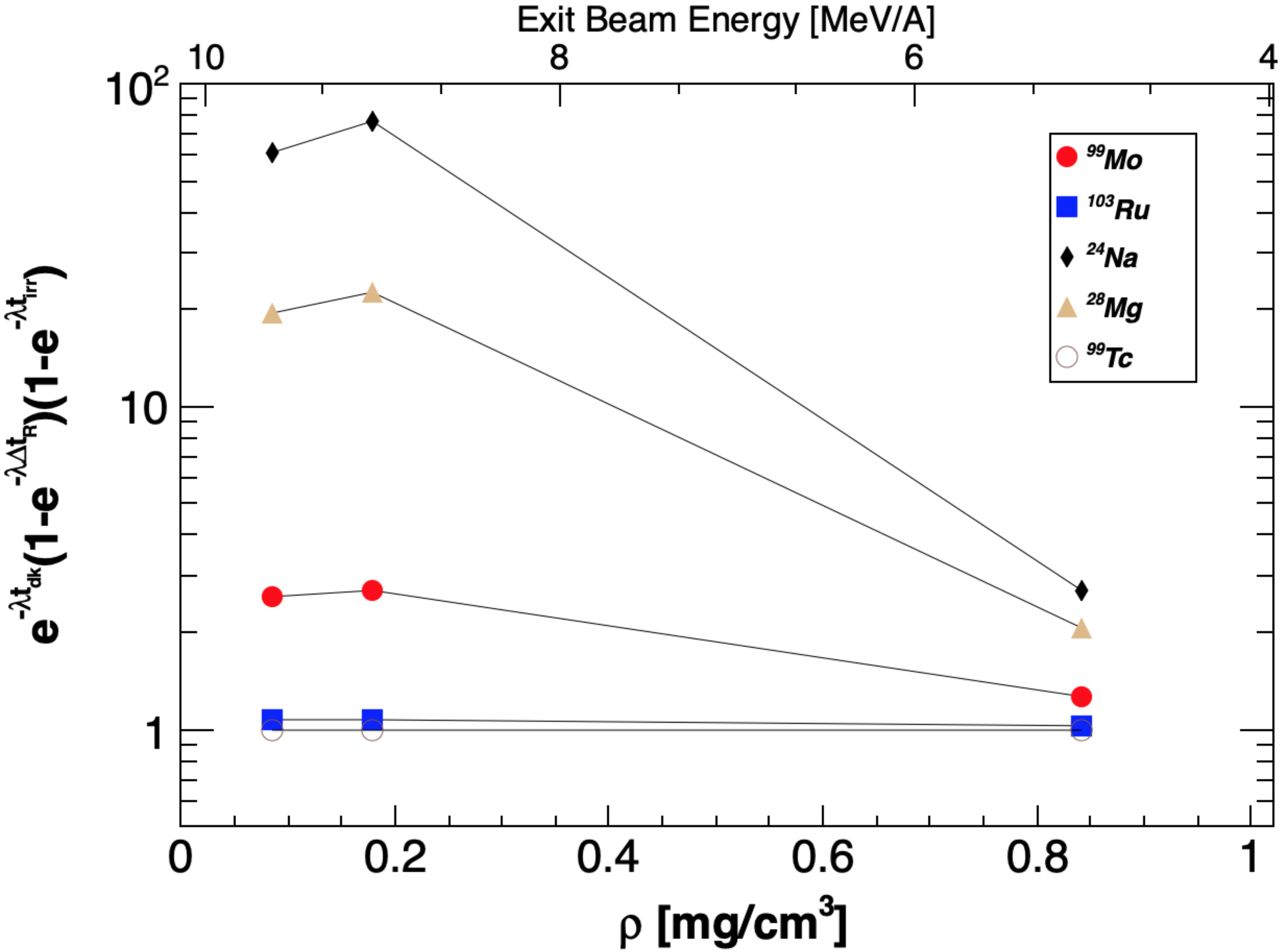


Medical Isotopes: Metals offer many options



● PET
● SPECT
● Beta Therapy
● Alpha Therapy
● Auger e⁻ Therapy

T.I. Kostelnik and C. Orvig, Chem. Rev. 2019, 119, 902-956


Exit Beam Energy [MeV/A]
10
8
6
4
10^2
10
1
$e^{-\lambda t_{dk}}(1-e^{-\lambda \Delta t_R})(1-e^{-\lambda t_{irr}})$
0
0.2
0.4
0.6
0.8
1
ρ [mg/cm^3]
● ^{99}Mo
■ ^{103}Ru
♦ ^{24}Na
▲ ^{28}Mg
○ ^{99}Tc

Energy threshold and Q-value

For an interaction of projectile nuclei of mass m_1, and target nuclei of mass m_2, the Q-value (Q [MeV]) is defined as:

$$Q = m_1 \cdot c^2 + m_2 \cdot c^2 - \sum_i m_i \cdot c^2 \tag{2.13}$$

where m_i denotes the mass of i-th produced nucleus. Obviously, for elastic scattering, $Q = 0$. For other processes, two cases are possible:

- $Q < 0$ then the reaction is endo-energetic, and the final system has higher mass (that has to be delivered in the form of kinetic energy of colliding particles),
- $Q > 0$ then the reaction is exo-energetic, and the final system has lower mass.

The exo-energetic reaction may occur at any energy. For the endo-energetic reaction, a laboratory energy limit, called threshold energy (E_{thr}), sets the minimum energy needed for the reaction to occur. The laboratory energy threshold can be calculated from the conservation of momentum and energy to link it with Q-value:

$$E_{thr} = |Q| \cdot \left(1 + \frac{m_1}{m_2}\right) \tag{2.14}$$