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Investigation of the ^{99}Mo production via neutron capture ^{98}Mo(n,γ)^{99}Mo with a high-current accelerator-based neutron source

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Project Outline
Who? What? Why?

- FH Aachen University of Applied Sciences
  - Department of Energy Technology
- University of Cologne
  - Faculty of Mathematics and Natural Sciences, Department of Nuclear Chemistry
  - Faculty of Medicine and University Hospital Cologne, Institute of Radiochemistry and Experimental Molecular Imaging
- Leibniz University of Hannover
  - Institute of Radioecology and Radiation Protection
- Jülich Research Center
  - Jülich Center for Neutron Science (JCNS) Institute High Brilliance Neutron Source (HBS)
  - Institute of Neuroscience and Medicine (INM-5) – Nuclear Chemistry

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Project Outline
Who? **What?** Why?

Aim: Production of ^{99}Mo-based radiodiagnostics using an accelerator based neutron source

Sub-projects:

1. Neutron Target Technology

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Developing high neutron flux density neutron target technology is crucial for irradiation with reduced radiation doses, ensuring safe handling and processing of Mo samples post-irradiation

2. Radiation Protection and Disposal

Leibniz Universität Hannover

Addressing safety concerns, this sub-project aims to determine radiation protection and disposal issues pertinent to the novel ^{99}Mo production process, ensuring a secure and sustainable approach

3. Process Optimization

University of Cologne

This involves refining the processes for generating ^{99}Mo-based radiodiagnostics, as well as improving their processing and utilization in clinical settings

Project Outline
Who? What? **Why?**

Key radioisotope : ^{99m}Tc (Technetium-99m)

- One of the most commonly used radioisotope for diagnostics ($t_{1/2}$ = 6h)
- **Produced from ^{99}Mo ($t_{1/2}$ = 66h)**
- Around 80% of nuclear medicine diagnostic exams globally use ^{99m}Tc (40 million per year), with 25% in Europe [1]
- In Germany, approximately 60,000 diagnostic exams per week, consuming nearly 10% of the global annual ^{99m}Tc requirement [2]

Conventional ^{99}Mo Production:

- ^{99}Mo is conventionally produced by fission of ^{235}U in reactors with high neutron flux [3]
- Generation of significant radioactive waste requiring disposal and non-proliferation measures [3]

Supply Chain Impact:

- Due to limited half-life, disruptions in the supply chain impact medical tests, patient treatment, and health
- Cancellation or delay of important medical tests is common without a stable supply of isotopes

Subproject 1
Neutron Target Technology

Aim: Design concept for a Target-Moderator-Reflector system for the production of ^{99}Mo isotope and its handling and transport

1. Development of a concept for ^{99}Mo production

   - Experimental validation of simulation codes such as PHITS and FLUKA
     - **Experiments at the FH Aachen irradiation chamber**
   - Benchmarking of the neutron moderator and reflector system
     - **Experiment at the JULIC platform**

2. Development and validation of a high-performance neutron target

3. Development of an automatic handling and transport system for ^{99}Mo-irradiated samples

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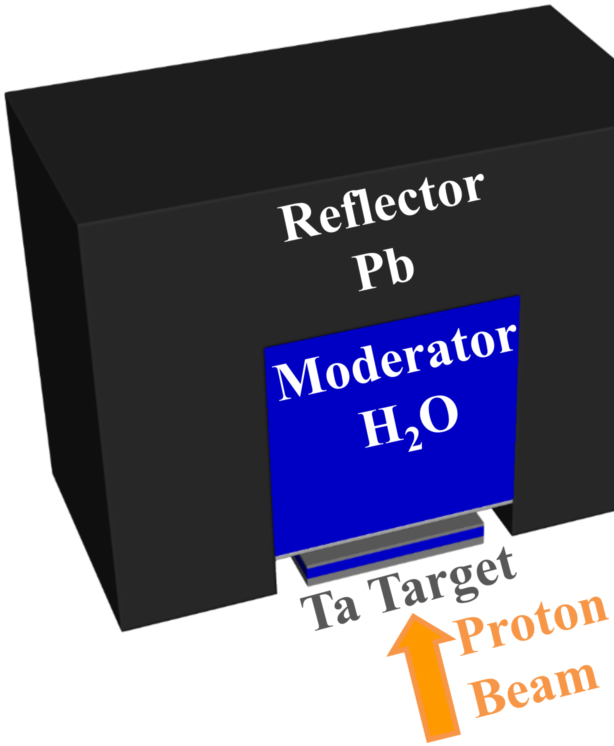
Subproject 1
Method

- Fast neutron generation
- 70 MeV, 100 mA proton current beam
- Tantalum target
- Neutron moderation
- Neutron reflection
- Activation of natural Mo sample
  - Thermal neutrons (σ = 0.13 b)
  - Epithermal neutrons (σ = 6.5 b)
- PHITS Simulations
- Experiments for validation of PHITS



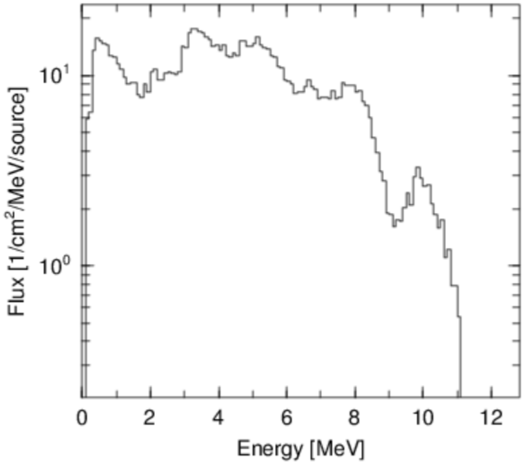
Experiment
Irradiation at FH Aachen irradiation chamber

Irradiation chamber:

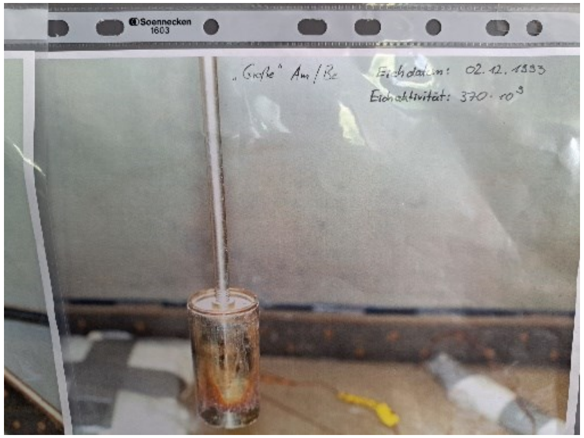
- 5 plexiglass tubes
- Paraffin wax moderator
- Iron – Cadmium – Iron cover

AmBe neutron source:

- Calibration date: 02.12.1993
- Calibration activity: 10 Ci
- Activity: 9.7 Ci
- Yield: $2.2 \pm 0.2 * 10^7$ s^{-1}



Neutron flux spectrum of the AmBe source



Americium – Berylium source

Irradiation Chamber

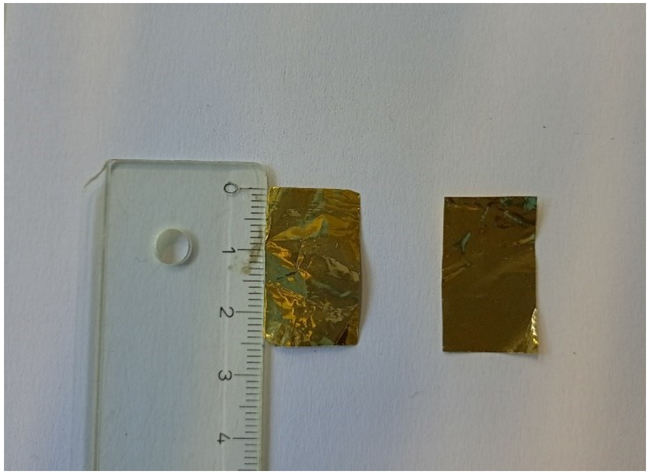
Experiment
Sample preparation

Au sample:

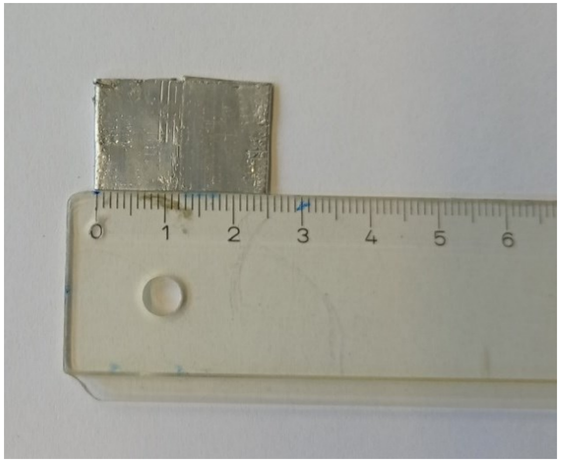
- 2.5 x 1.5 x 0.0025 cm
- 0.1648 ± 0.003 g

Au + Cd sample:

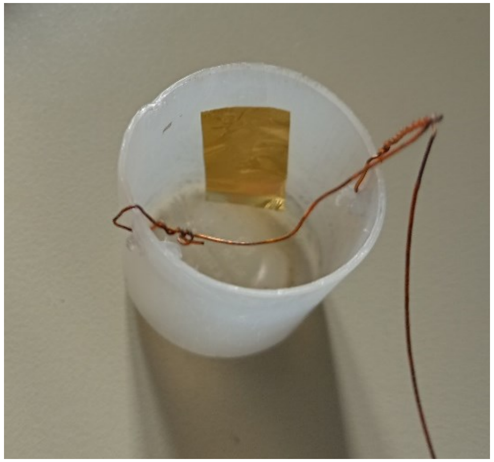
- Au: 2.5 x 1.5 x 0.0025 cm
  0.1656 ± 0.003 g
- Cd: 5.2 x 1.6 x 0.05 cm
  3.5093 ± 0.07 g

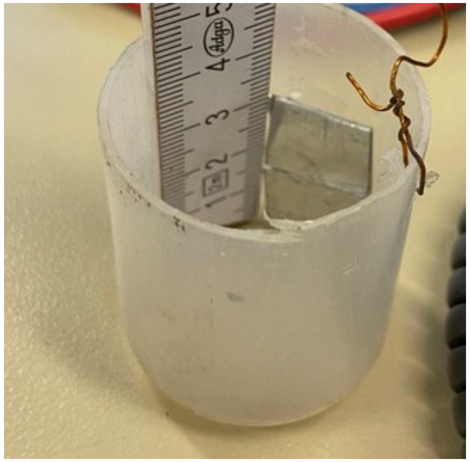


Gold foil



Gold foil wrapped with Cd





Samples in a polythene cup

Experiment
Neutron irradiation at Big Karl

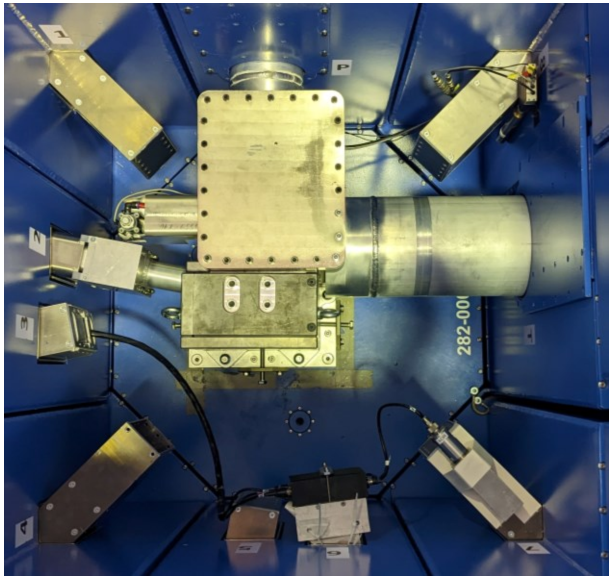
Irradiation @ Big Karl:

- 45 MeV protons
- Average current: 35 nA
- 3-day irradiation

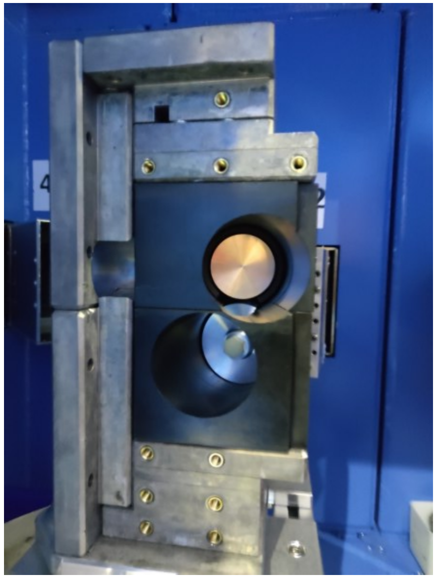
Sample dimensions:

- Natural Mo: 2.5 x 2.5 x 0.1 cm
- Au: 1.5 x 1.5 x 0.003 cm

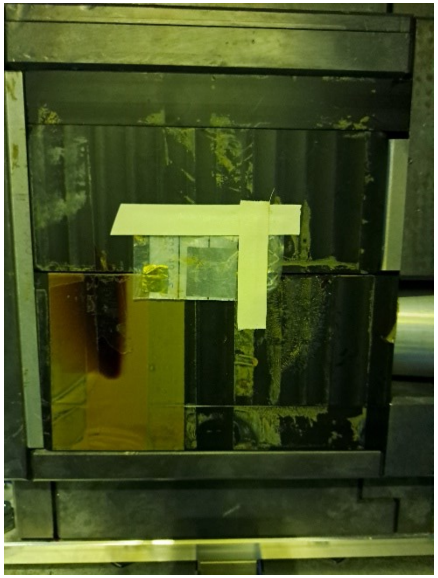
Target station



View from top of the TMR



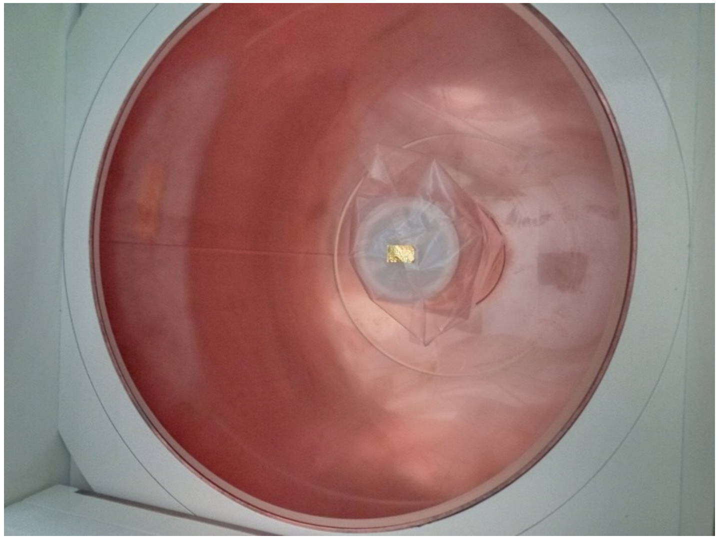
Side view of the TMR



Sample position

Experiment
Gamma spectrum measurement

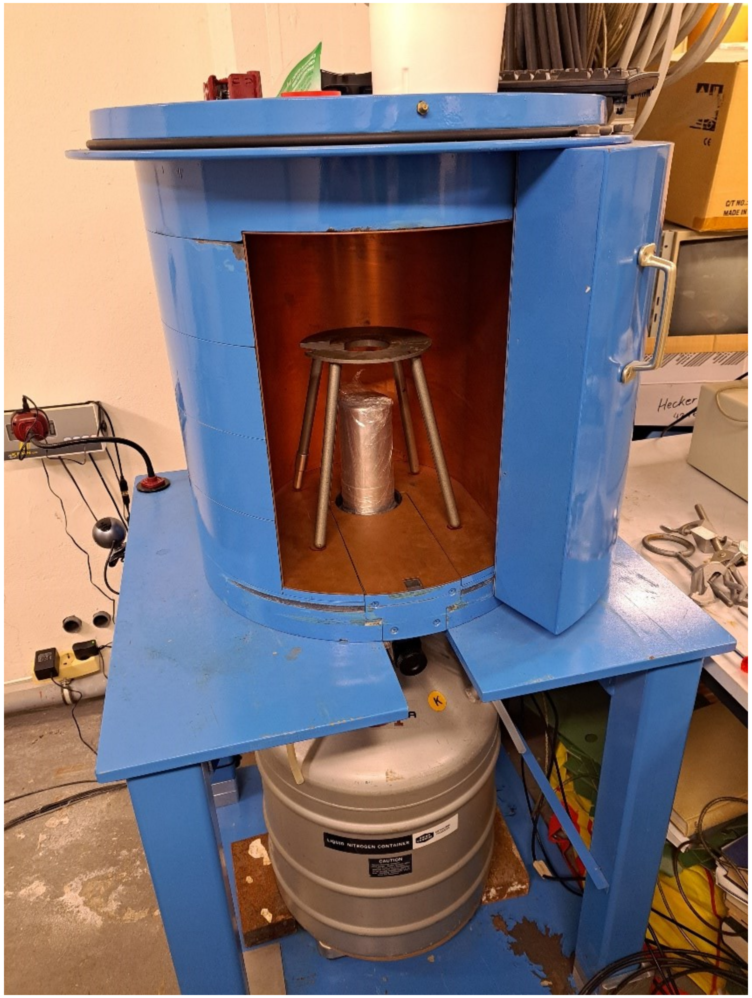
Measurement with HPGe-Detector at FH Aachen



Detector Chamber

Detecor set-up

Measurement with HPGe-Detector at Big Karl



Simulations
Simulation codes and data post-processing

PHITS

- Simulation of neutron flux
- Simulation of proton beam
- Neutron flux within the foil sample
- Induced activity (D-Chain)

Python

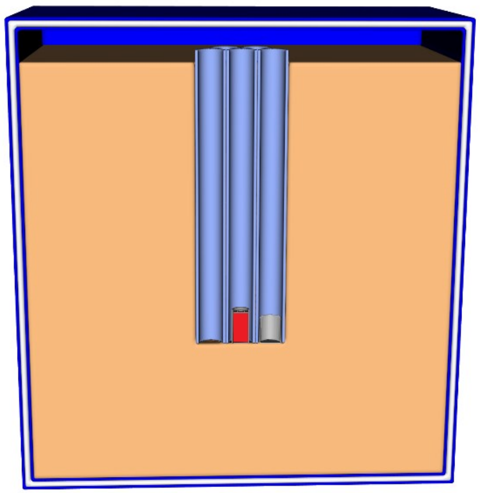
- Data extraction from output files
- Activity calculation
- Data analysis

$$A = \frac{m}{M} N \cdot h \cdot (\sigma_{th} \cdot \phi_{th} \cdot G_{th} + I_{\gamma} \cdot \phi_{epi} \cdot G_{epi}) \cdot \left(1 - e^{-\frac{ln2 \cdot t_b}{t_{1/2}}}\right)$$ Neutron flux method

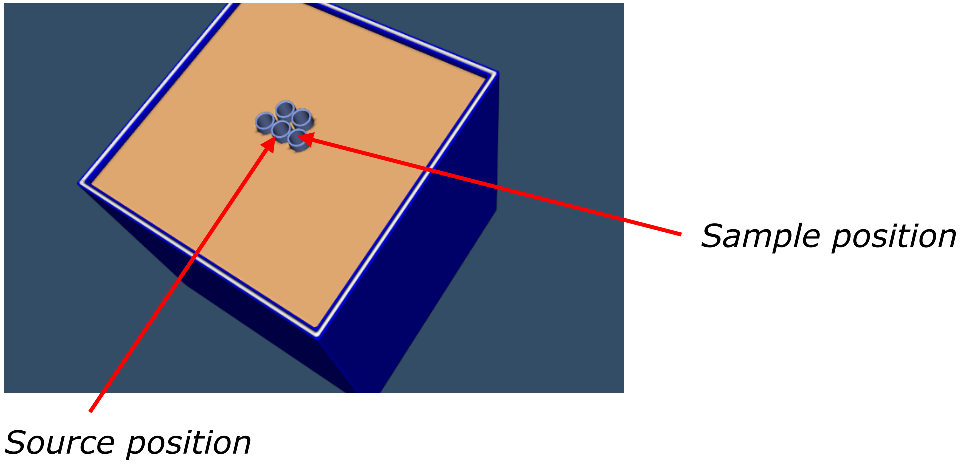
$$A = \left(N_0 \cdot \left(\sum_i^n \sigma_i(E) \cdot \phi_i(E)\right) \cdot t \cdot \lambda\right) - \left[\left(N_0 \cdot \left(\sum_i^n \sigma_i(E) \cdot \phi_i(E)\right) \cdot t \cdot \lambda\right) \cdot \left(1 - e^{-\lambda \cdot t}\right)\right]$$ D-Chain method

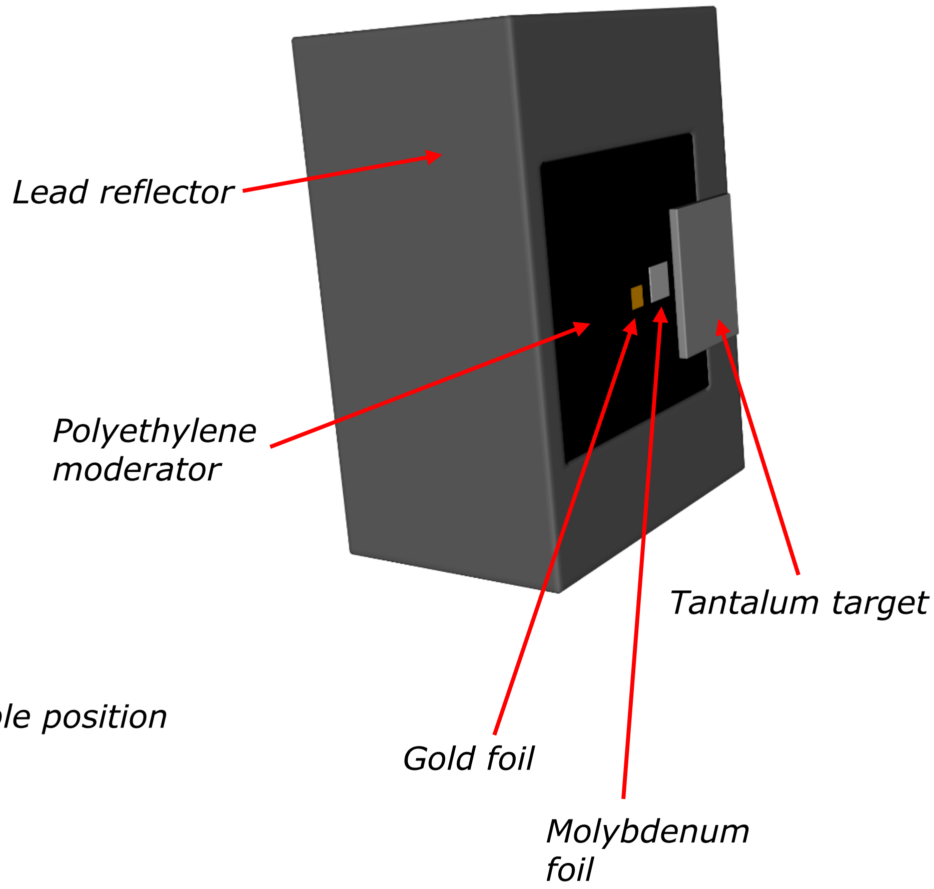
m − mass of the sample (grams)
h − aboundance of 98Mo (0.2419 for natural Mo)
σ_{th} − thermal cross − section for (n, γ) reactions
ϕ_{th} − thermal neutron flux
G_{th} − thermal neutron self shielding factor
I_{γ} − radiative neutron capture resonance integral
ϕ_{epi} − epithermal neutron flux
G_{epi} − epithermal neutron self shielding factor
$t_{1/2}$ − half − life of 99Mo
t_b − irradiation time

Simulations
Reproduction of experimental conditions



Cut through the y=0 plane of the irradiation chamber





Experiment and Simulation
Preliminary Results

$$A = \left(N_0 \cdot \left(\sum_i^n \sigma_i(E) \cdot \phi_i(E)\right) \cdot t \cdot \lambda\right) - \left[\left(N_0 \cdot \left(\sum_i^n \sigma_i(E) \cdot \phi_i(E)\right) \cdot t \cdot \lambda\right) \cdot \left(1 - e^{-\lambda \cdot t}\right)\right]$$

$$A = \frac{m}{M} N \cdot h \cdot \left(\sigma_{th} \cdot \phi_{th} \cdot G_{th} + I_\gamma \cdot \phi_{epi} \cdot G_{epi}\right) \cdot \left(1 - e^{-\frac{ln2 \cdot t_b}{t_{1/2}}}\right)$$

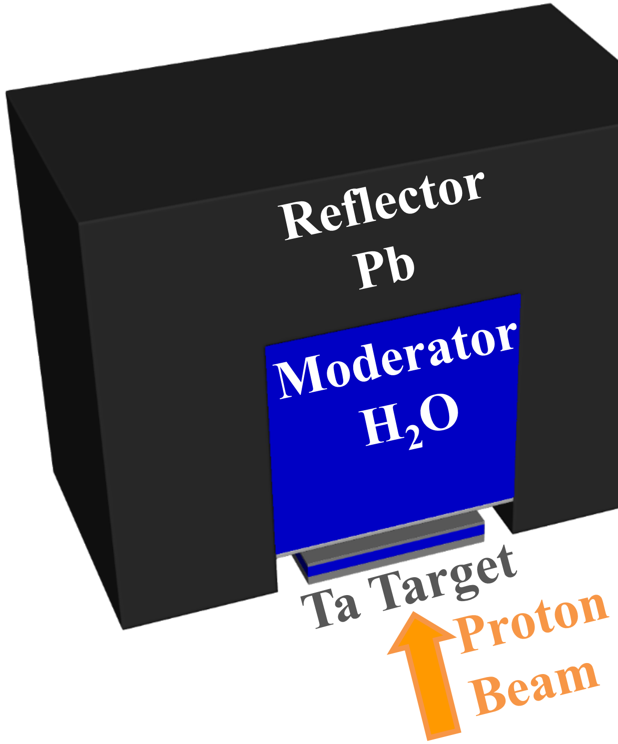
	Activity [Bq]	
	With Cd	Without Cd
Experiment	108±10	238±23
Flux analysis	125±15	264±27
D-chain tally	86	227

Table 1. *Experimental results from the irradiation at the FH Aachen irradiation chamber compared to simulations results*

Optimization strategies

Optimization of following parameters to achieve an activity of 2400 Ci of ^{99}Mo:

- Design of the moderator and reflector to maximize the integral epithermal flux
- Moderator material
  - H2O, D2O, ZrH, LiF
- Reflector material
  - Pb, MgO, Be, Fe



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