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PERSPECTIVES ON THE PRODUCTION OF ^{99}MO IN THE FRAMEWORK OF IFMIF/ELAMAT PROJECT

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Context - $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$

General considerations

^{99}Mo – Neutron production routes


Calculation methodology and assumptions

Results

Conclusions

- $^{99\text{m}}\text{Tc}$ is worldwide used in more than 80% of nuclear medicine procedures
 - Short half-live (6 hours)
 - Production mainly based on the radioactive decay of ^{99}Mo ($T_{1/2}=66$ hours)

 - Production of ^{99}Mo based on fission method in nuclear research reactors
 - 5 reactors represent more than 90% of the total production
 - These reactors are more than 40 years old

 - Medical isotopes crisis occurred in 2009
 - Shutdown of Petten in Netherland
 - Shutdown of Chalk river in Canada
-  **Serious concern on ^{99}Mo supply**
- International agencies (IAEA, OECD) recommendations:
 - Increasing of diversity and redundancy of ^{99}Mo supply (especially small & medium scale facilities)
 - Conversion technology: HEU -> LEU (security & non-proliferation)

Competitiveness of accelerator-based neutron source for the production of ^{99}Mo using non-fission method ?

■ World demand:

- ^{99m}Tc : 160000 doses per day, ~25 mCi per doses, namely ~4000 Ci per day
- ^{99}Mo : 10000 6-days Ci per week

6-days Ci: Activity unit in Ci, 7 or 8 days after the end of irradiation
(transportation, sample processing)

■ Producer facility size for the production of ^{99}Mo :

- Small : < 7400 6-days GBq / week
- Medium : 7400 6-days GBq / week -> 37000 6-days GBq / week
- Large : > 37000 6-days GBq / week

■ Radioisotopic purity:

- Final product administrated to patients
- Radiological contaminants as low as possible

Keys parameters studied: total activity, specific activity, radioisotopic purity

■ Two reactions in case of non-fission method, based on Mo sample:

- $^{98}\text{Mo}(n,\gamma)^{99}\text{Mo}$
- $^{100}\text{Mo}(n,2n)^{99}\text{Mo}$

■ $^{98}\text{Mo}(n,\gamma)^{99}\text{Mo}$:

- Thermal & epithermal neutrons
- Already used in nuclear research reactors

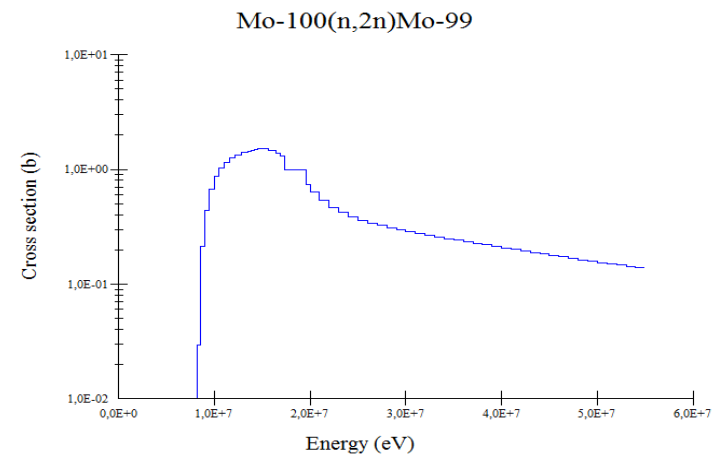
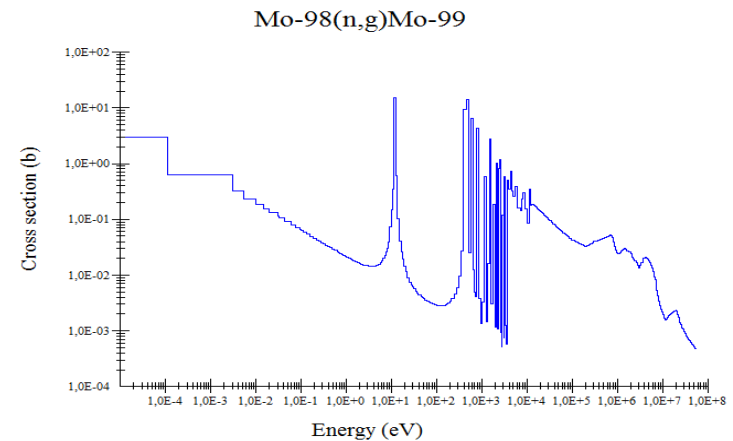
Requirements for accelerator-based neutron source:

- Thermal neutron flux $> 10^{14} \text{ cm}^2 \cdot \text{s}^{-1}$
- Moderator

■ $^{100}\text{Mo}(n,2n)^{99}\text{Mo}$:

- High-energy neutrons (threshold $\sim 8\text{MeV}$)
- Maximum cross section $\sim 14 \text{ MeV}$

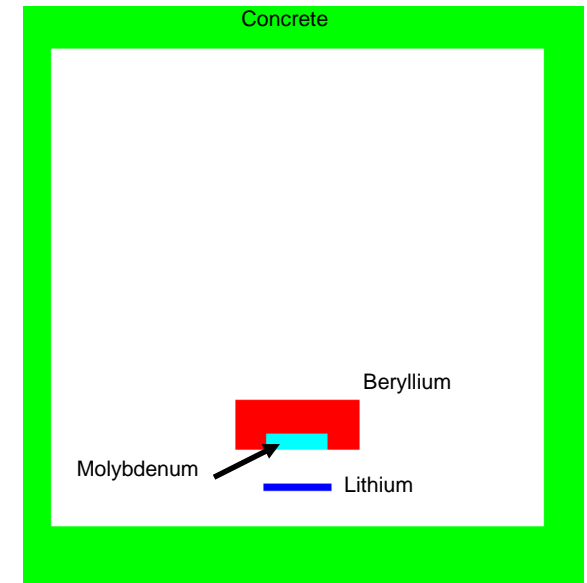
↳ This fits perfectly with IFMIF/ELAMAT neutron source characteristics



- Transport code: MCNPX 2.7.b
 - Neutron scattering library: ENDF/B-VII.1
 - Deuteron/Lithium interaction: Bertini model

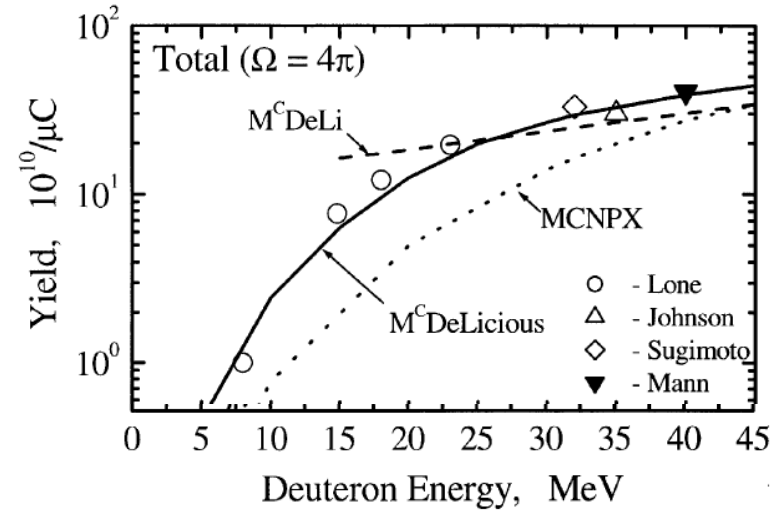
- Activation code: FISPACT 2007 (EAF 2007 neutron library)

- Assumptions (simplified geometry):
 - Beam:
 - Intensity: 125 mA
 - Energy: 40 MeV
 - Footprint: 50 mm x 200 mm
 - Mo sample:
 - 50 mm x 200 mm
 - 10 mm thick (470 grams for MoO₃, 1022 grams for ^{met}Mo)
 - Localisation: 10 cm behind Lithium target (keep place for irradiation module)
 - Moderator: Beryllium, 100 mm thick
 - Activation calculations:
 - No impurities considered
 - Irradiation time: 6 days (weekly production)
 - Cooling time: 8 days (sample processing)

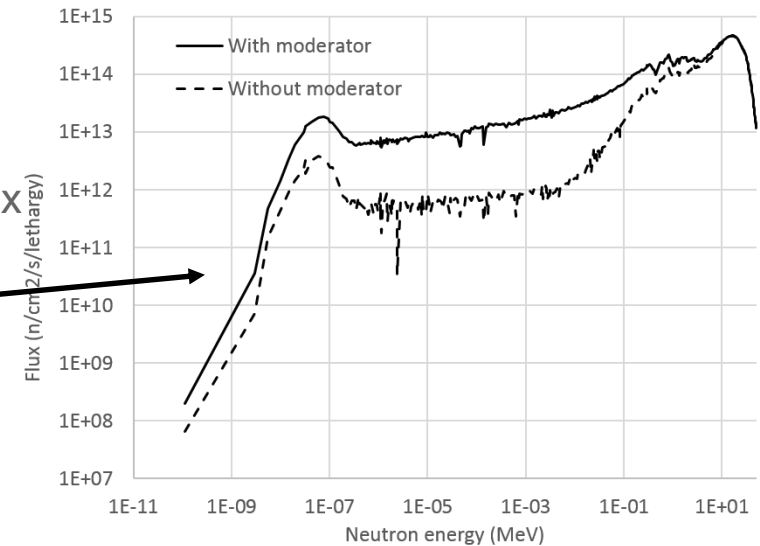
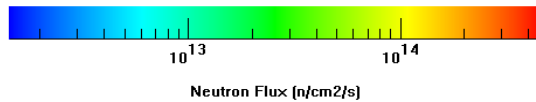
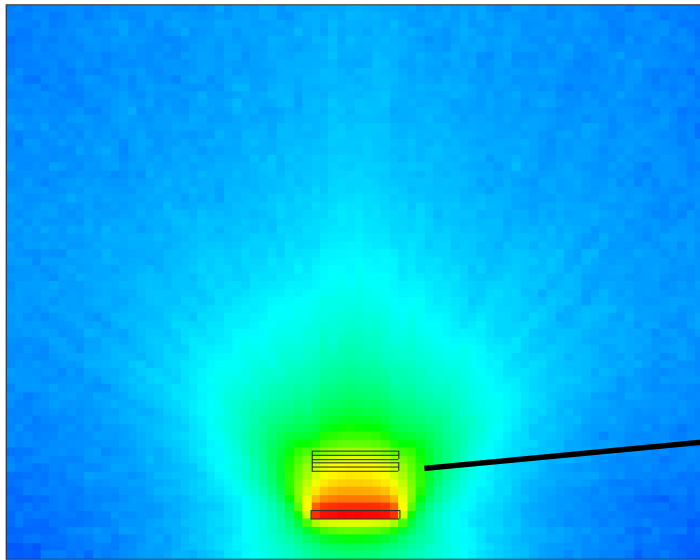


- Neutron rate in 4Pi:
 - Simulation: $3.75 \cdot 10^{11}$ neutrons. μC^{-1}
 - Experiment: $4 \cdot 10^{11}$ neutrons. μC^{-1}

- Strong enhancement of thermal (x4) and epithermal (x10) neutrons with Beryllium moderator (10 cm thick)

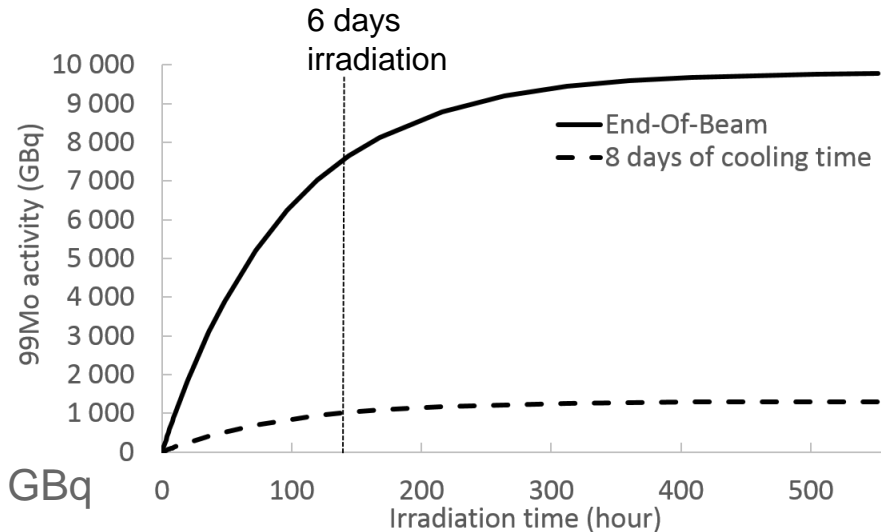


Simakov, J. Nuc. Mat. 307-311 (2002), pp. 1710-1714



- Mo sample:
 - Natural isotopic abundance
 - Oxide form
 - 470 grams (10 mm thick)
 - Without moderator

- After 6 days irradiation, ⁹⁹Mo activity:
 - T0: 7650 GBq
 - 8 days cooling time: 1000 6-days GBq



Only 27% of the median value for the small scale facility (3700 6-days GBq)
91% from ¹⁰⁰Mo(n,2n)⁹⁹Mo

- Fulfill the facility size requirements:
 - Increasing of sample mass
 - Use of high-enriched sample (⁹⁸Mo, ¹⁰⁰Mo)

Sample masses (kg) to fulfil facility lower limit

Scale facility	^{nat} MoO ₃	⁹⁸ MoO ₃	¹⁰⁰ MoO ₃
small (*)	1.71	4.61	0.19
medium	3.42	9.22	0.37
large	17.1	46.1	1.86

* mean value

Specific activity: 2.1 GBq/g ^{nat}Mo, 0.8 GBq/g ⁹⁸Mo, 20 GBq/g ¹⁰⁰Mo

- Mo sample with moderator:
 - Natural abundance
 - Oxide form
 - 470 grams (10 mm thick)
 - Be moderator (10 cm thick)

Sample masses (kg) to fulfil facility lower limit

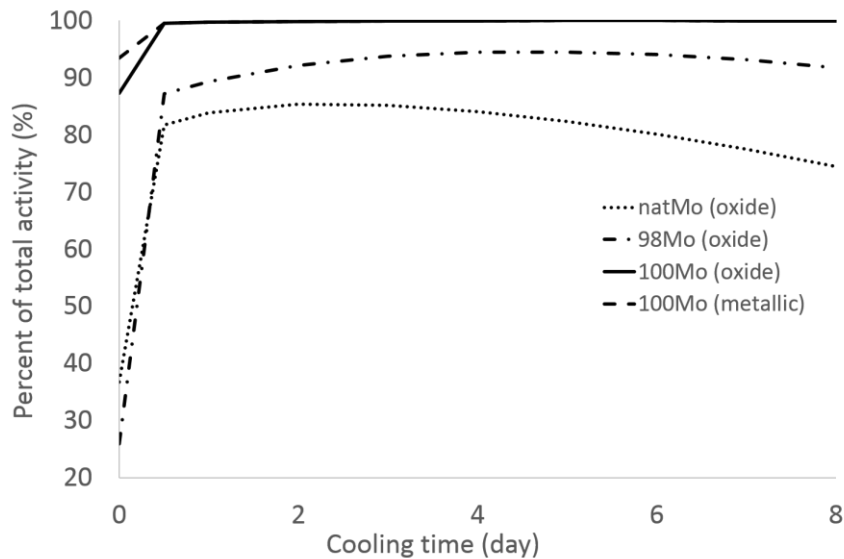
Scale facility	^{nat} MoO ₃	⁹⁸ MoO ₃	¹⁰⁰ MoO ₃
small	1.13	0.71	0.18
medium	2.26	1.42	0.37
large	11.3	7.11	1.83

Effect on ^{nat}Mo and ⁹⁸Mo due to enhancement of ⁹⁸Mo(n,γ)⁹⁹Mo contribution

- With metallic sample instead of oxide: reduction of sample masses by 2/3
 - Small facility (3700 6-days GBq /week): 125 grams of ¹⁰⁰Mo
 - Medium facility (7400 6-days GBq /week): 250 grams of ¹⁰⁰Mo
 - Large facility (37000 6-days GBq /week): 1250 grams of ¹⁰⁰Mo

- With sample characteristics:
 - 200x50x10 mm³
 - 10 cm behind Li target
 - Mass:1022 grams

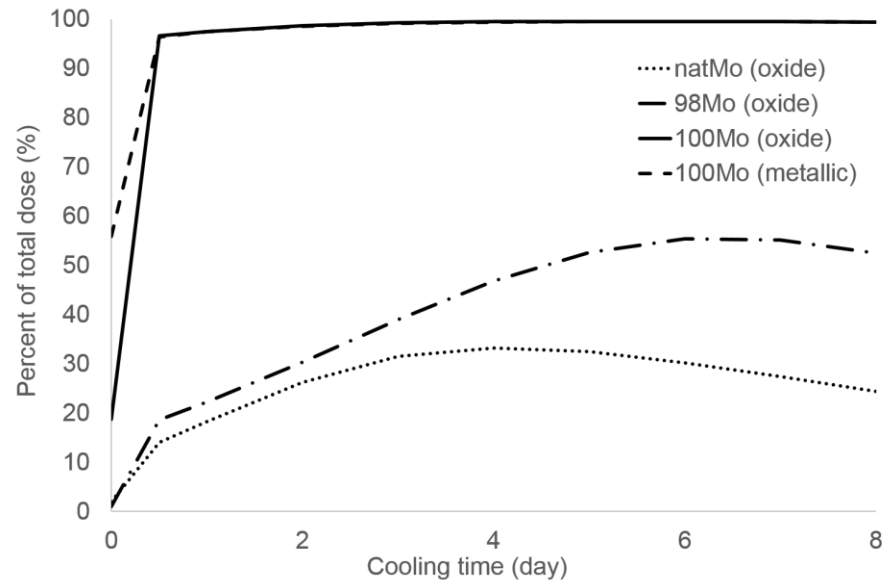
➔ **Medium scale facility**



Only ^{100}Mo sample provides high radioisotopic purity. Sample processing needed?

In other case ($^{\text{nat}}\text{Mo}$ and ^{98}Mo), extraction of contaminants are needed if possible.

- $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ contribution to the total activity and the total dose:
- ^{98}Mo and $^{\text{nat}}\text{Mo}$: large contribution of Nb isotopes to total dose rate (main contributor $^{92\text{m}}\text{Nb}$ after 4 days of cooling time)
- ^{100}Mo sample: more than 99% in activity (12 hours) and dose (3 days)



- ^{99}Mo activity evaluated in the framework of IFMIF/ELAMAT project
 - Sample dimensions: 200x50x10
 - 10 cm behind Lithium target & irradiation module
 - 470 grams for oxide form, 1022 grams for metallic form

- Advantages:
 - Not the primary goal of the facility
 - Neutron activation: sample mass adjustment if available space

- ^{100}Mo sample is the most efficient route for the production of ^{99}Mo (specific activity: 2.1 GBq/g $^{\text{nat}}\text{Mo}$, 0.8 GBq/g ^{98}Mo , 20 GBq/g ^{100}Mo , oxide form, without moderator)
 - Small scale facility: 190 grams of $^{100}\text{MoO}_3$ (125 grams for $^{100}\text{Mo}_{\text{met}}$)
 - Medium scale facility: 380 grams of $^{100}\text{MoO}_3$ (250 grams for $^{100}\text{Mo}_{\text{met}}$)
 - Large scale facility, 1800 grams of $^{100}\text{MoO}_3$ (1250 grams for $^{100}\text{Mo}_{\text{met}}$)

- ^{100}Mo sample provides a high radioisotopic purity compared to $^{\text{nat}}\text{Mo}$ & ^{98}Mo
 - Sample processing needed for labelling for ^{100}Mo ?
 - Recycling of ^{100}Mo needed due to the expensive cost

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