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Robust and Scalable Production of Key Medical Radioisotopes (Mo-99, Lu-177, Ac-225) via Commercial Light Water Reactors

Naoyuki Takaki
Department of Nuclear Safety Engineering
Tokyo City University

東京都市大学
TOKYO CITY UNIVERSITY

Contents

Why fission reactor ?

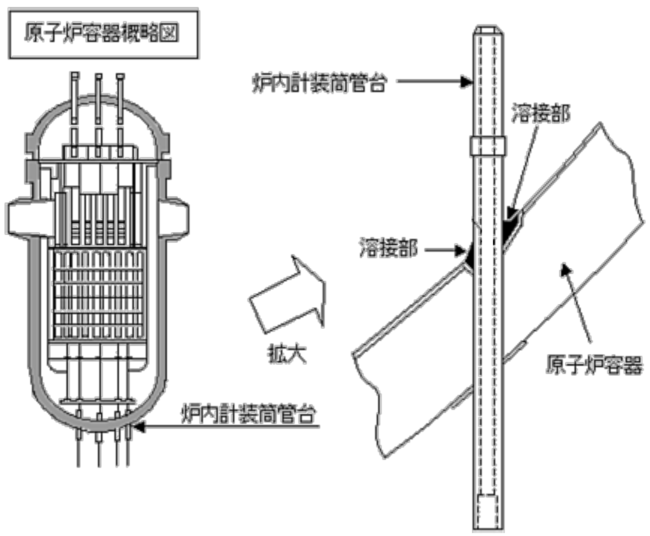
- High neutron flux (∵ $\Sigma_f \phi \propto P$)
- Large irradiation area (∵ large core volume)
- High heat removal capability (∵ cooling for nuclear fuel)
- Excellent energy balance (∵ Energy not consumed but produced)

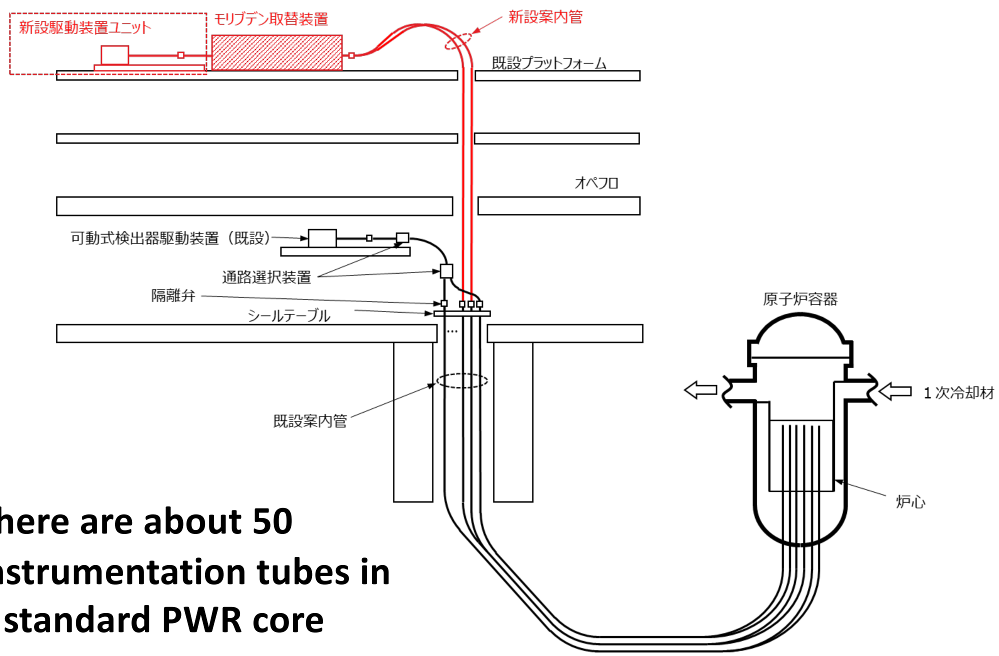
Why commercial LWR ?

- Stable supply (∵ stable operation with high capacity)
- High neutron fluence (∵ high capacity factor & longer cycle)
- No need for specialized facility (∵ existing plant owned by utilities)
- Fit for “full cost recovery” policy (∵ Almost all capital, fuel and O&M cost covered by electricity sales)

Target Loading Method into PWR for Mo/Tc and Lu-177 production

- Use instrumentation tube which penetrate the pressure vessel.
- The tube is originally equipped for movable in-core sensors (fission chambers) to detect axial neutron distribution once a month.





- There are about 50 instrumentation tubes in a standard PWR core

- For Mo/Tc production, 4 tubes are planned to use for 1 week irradiation.
- For Lu-177 production, 1 tubes for 2 week irradiation.

Mo Pellets and ^{99}Mo Production Rate

- Mo pellets are linked together by flexible connector
- The total length: 3600mm
- The total weight of Mo pellets in 4 strings : 520g (MoO_3: 780g)
- Enrichment of ^{98}Mo : 98%
  (^{98}Mo in natural Mo : 24.1%)

Production rate after 7 days irradiation

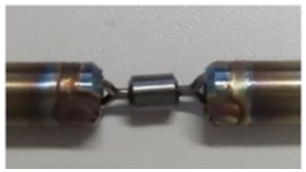
→ 275 GBq/g-^{98}Mo

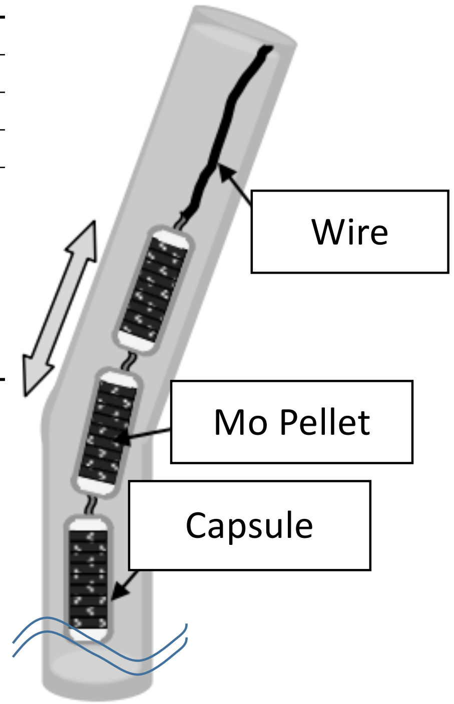
→ 600 [6-days calibrated Ci/week] [1)]

60% of annual demand (1000 Ci/week) in Japan

カプセル形状例

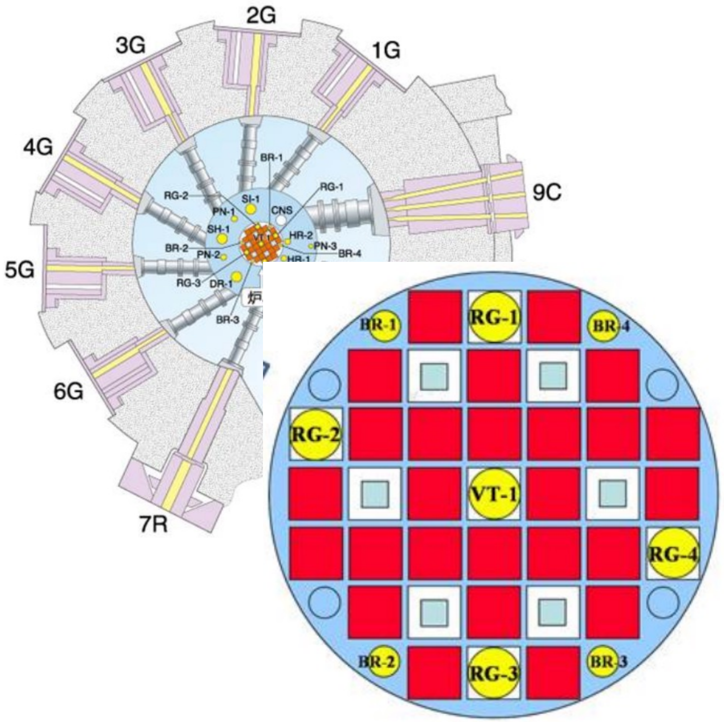
全　長	：約50mm
外　径	：約5mm
肉　厚	：0.5mm未満
材　質	：SUS304
連結方法	：スイベル構造

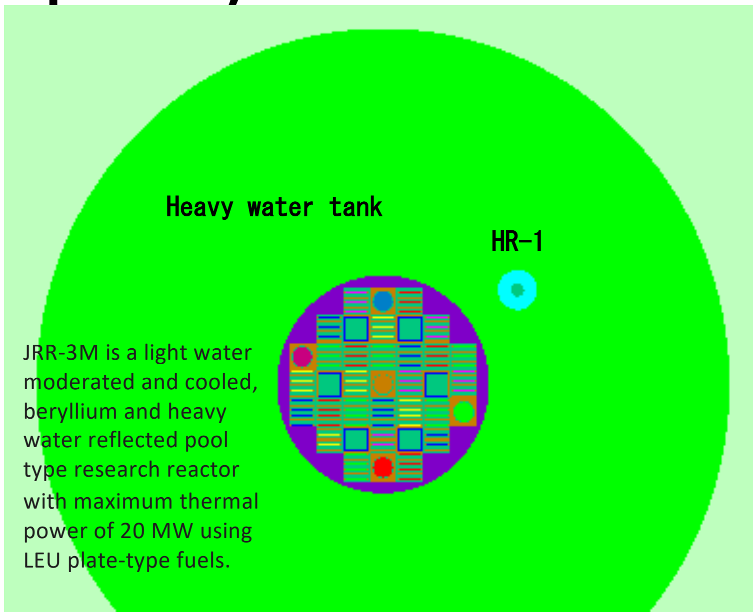




参考文献：
1. 高橋 静香、中原 隆、円谷 信一、小林 一太、那須 拓哉、高木 直行、「国内の原子力インフラを活用した医用RIの自給技術確立に向けた研究開発 n (3) PWRを用いたMo/Tc生成の概念検討」、日本原子力学会 2022年春の年会
2. 広瀬 彰、米田 政夫、木名瀬 政美、反田 孝美、和田 茂, JRR-3 を用いた 99Mo 製造に関する概念検討, JAEA-Technology, 2010-007

Lu-177 production experiment using JRR-3M (Aug.-Sep.2023)





JRR-3M is a light water moderated and cooled, beryllium and heavy water reflected pool type research reactor with maximum thermal power of 20 MW using LEU plate-type fuels.

Target material	Lu-176 (64.3% Enriched)
Mass	0.094mg
Irradiation position	HR-1
Neutron flux at HR-1	1.3×10^{14} [/cm^2・sec]
Irradition period	14days

Fig. JRR-3M and the core

Target capsule and PIE

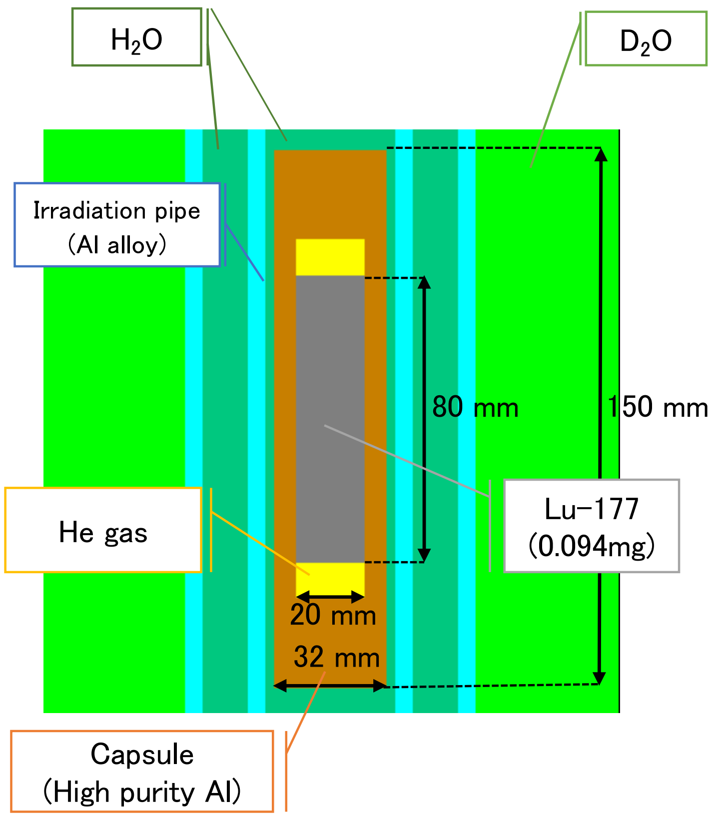


Fig. Target Capsule

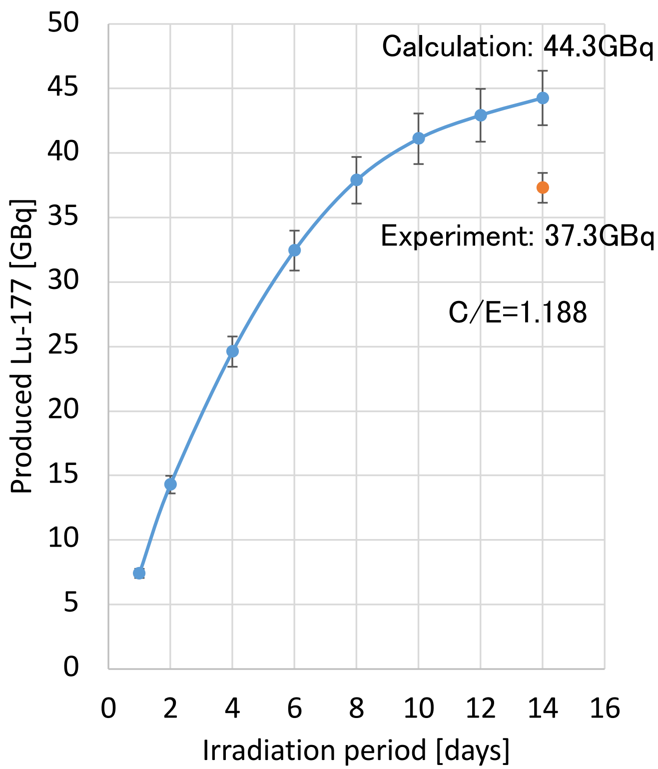
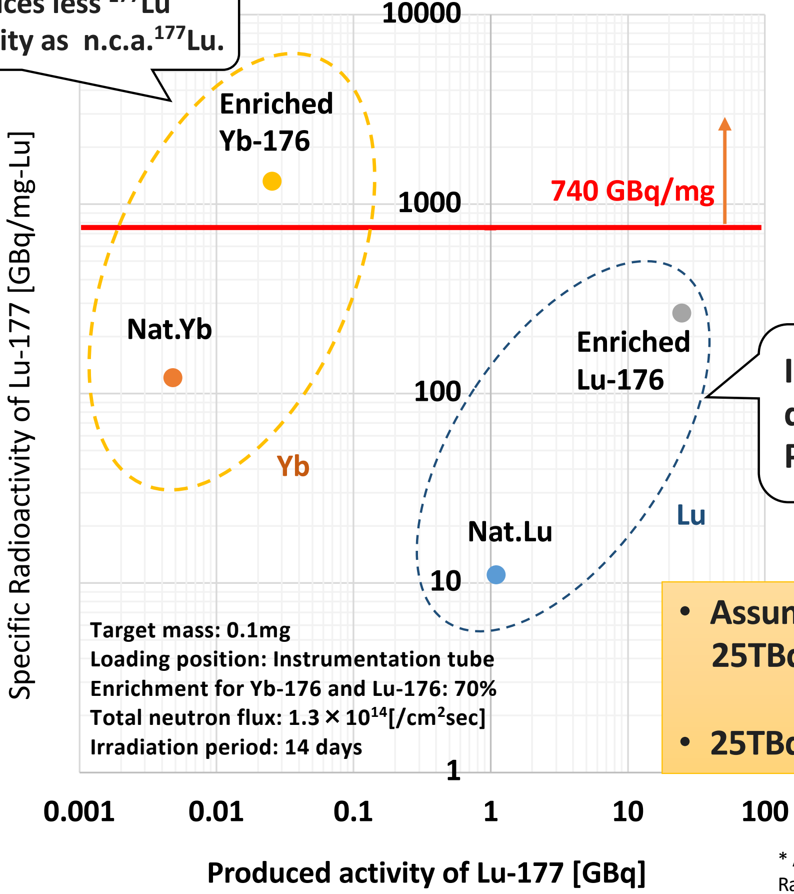


Fig. Lu−177 production in JRR−3M

Estimated production of Lu-177 in commercial PWR

Yb irradiation produces less ^{177}Lu but satisfies the purity as n.c.a.^{177}Lu.



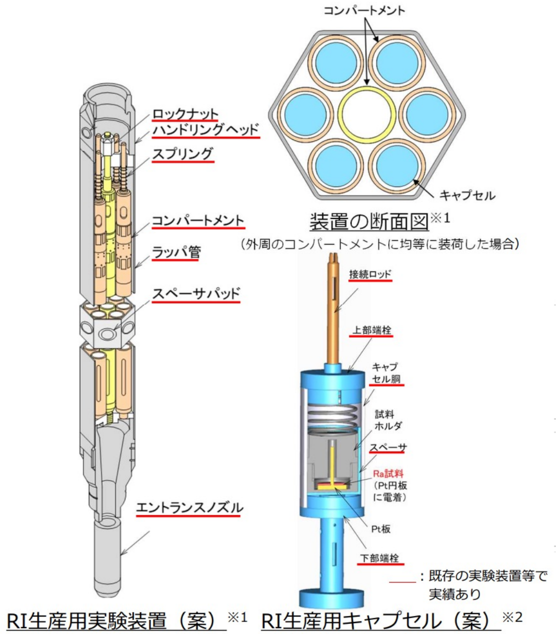
Peptide Receptor Radionuclide Therapy (PRRT) with ^{177}Lu-DOTA-peptides requires ^{177}Lu with high specific activity (SA) and values >740 GBq ^{177}Lu/mg-Lu*.

Irradiation of Lu gains larger amount of ^{177}Lu but does not meet the criteria of specific activity under PWR irradiation condition even using enriched ^{177}Lu.

- Assuming 100g of ^{176}Yb target, 25TBq of ^{177}Lu produced by 14 days irradiation.
- 25TBq × 24 irrd./y = 600TBq/y (=81,100doses =13,500patients)

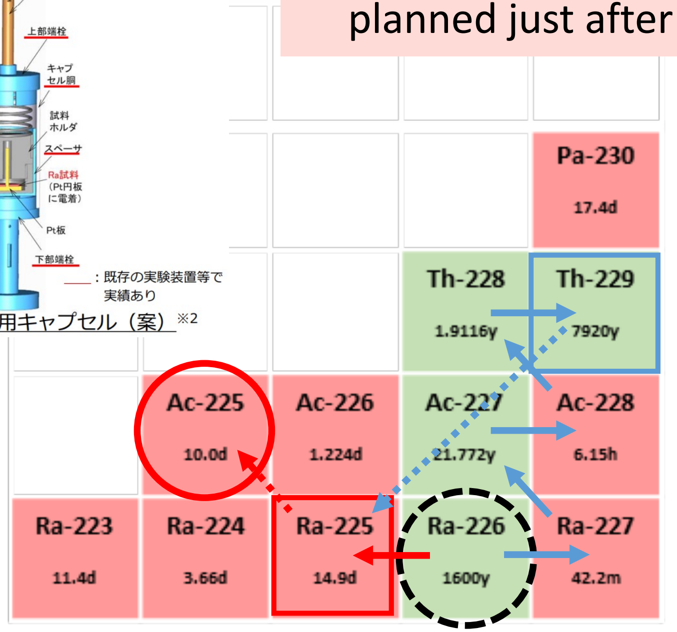
* A. Dash, M. R. A. Pillai, Furn F. Knapp Jr, "Production of 177Lu for Targeted Radionuclide Therapy: Available Options," Nucl Med Mol Imaging, 49, 85-107 (2015)

^{225}Ac production methods in fission reactors



Fast neutron path： ^{226}Ra(n,2n)^{225}Ra→^{225}Ac

- Joyo has potential to efficiently produce Ac-225 by this path[1)]. (about 60GBq/y by 60days×3times/y irradiation)
- Demonstration of Ac-225 production (by 10mg Ra-226 target) is planned just after its restart planned in 2026.



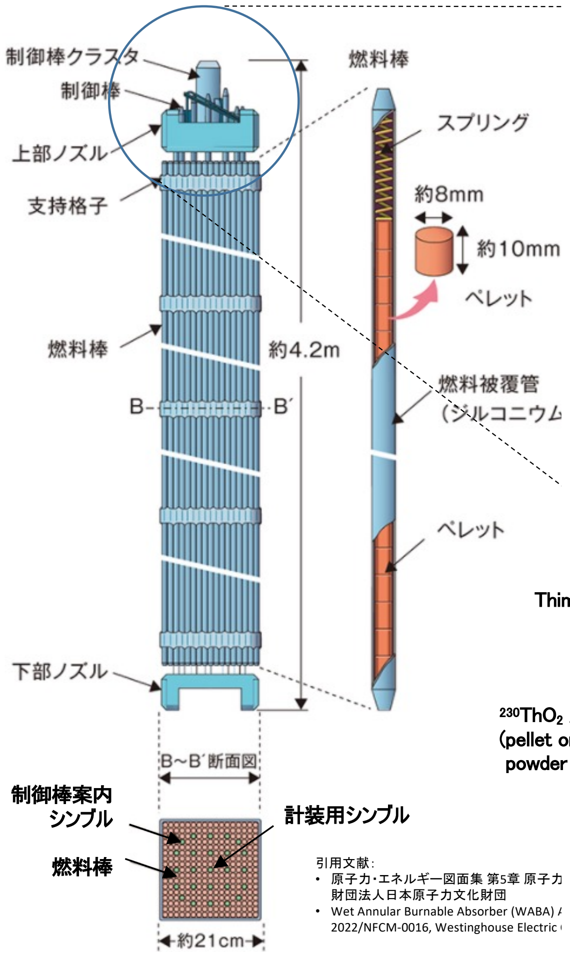
Thermal neutron path：

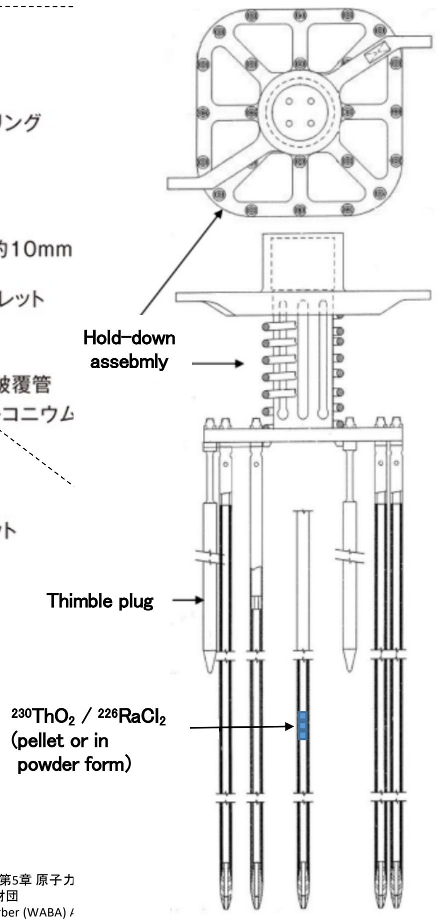
^{226}Ra(3n,2β)^{229}Th→^{225}Ra→^{225}Ac

- Commercial LWRs are applicable to produce Th-229 which is a permanent cow nuclide of Ac-225[2)].

1. Daiki Iwahashi *,Kota Kawamoto,Yuto Sasaki andNaoyuki Takaki, "Neutronic Study on Ac-225 Production for Cancer Therapy by (n,2n) Reaction of Ra-226 or Th-230 Using Fast Reactor Joyo", Processes 2022, 10(7), 1239; https://doi.org/10.3390/pr10071239
2. Daiki Iwahashi *,Yuto Sasaki,Tomoatsu Shinohara andNaoyuki Takaki, "Semi-Permanent Mass Production of Ac-225 for Cancer Therapy by the (3n,x) Reaction in Pressurized Water Reactor", Processes 2024, 12(1), 83; https://doi.org/10.3390/pr12010083

^{225}Ac production in commercial PWRs

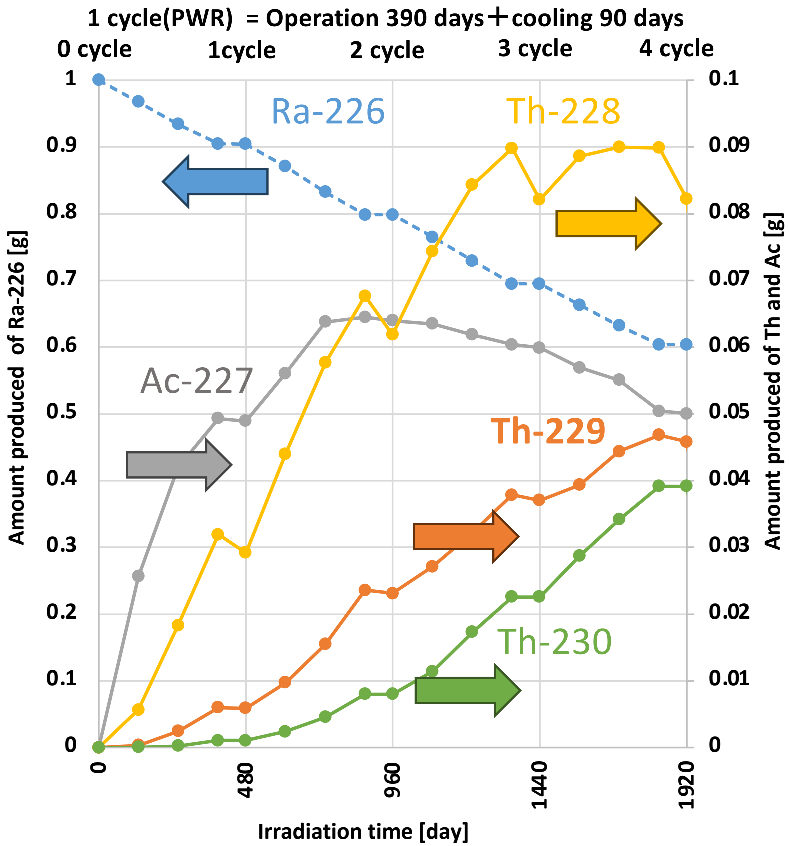




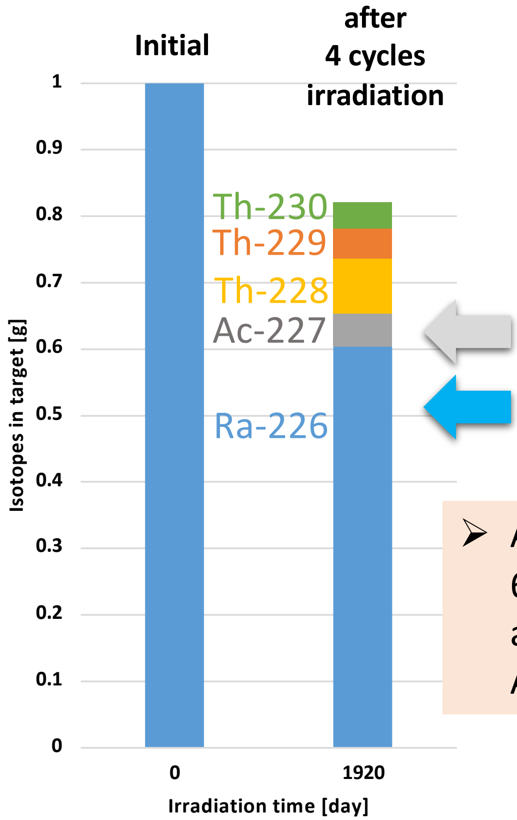
Loading method：

- One or several thimble plugs of hold down assembly are replaced by capsuled rods for irradiation.
- 1g of ^{226}Ra is loaded in pellet or powder form.
- The fuel assembly is shuffled 3 times and discharged after 4 cycle irradiation to the cooling pool as same as usual assemblies.

Th-229 production in PWR



Th-229 production through 4 cycle irradiation



Target composition

- After the irradiation, 60% of the Ra remains and newly produced Ac-227 is included

Indonesia Research Reactor (RSG-GAS)

First Working Core Configuration[3]

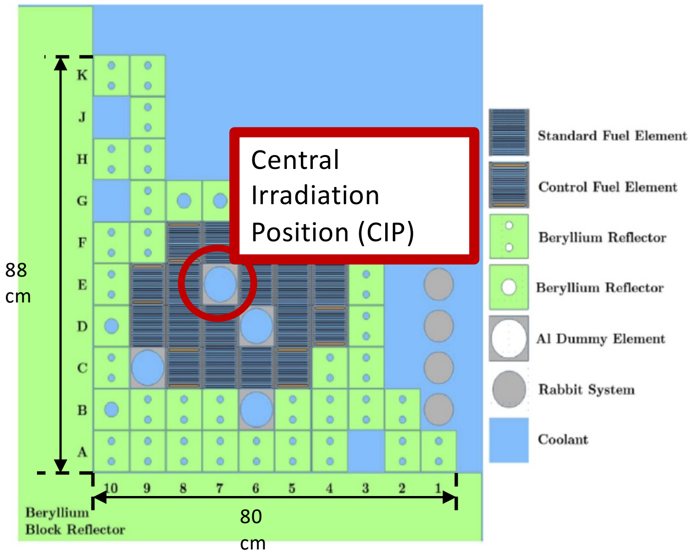


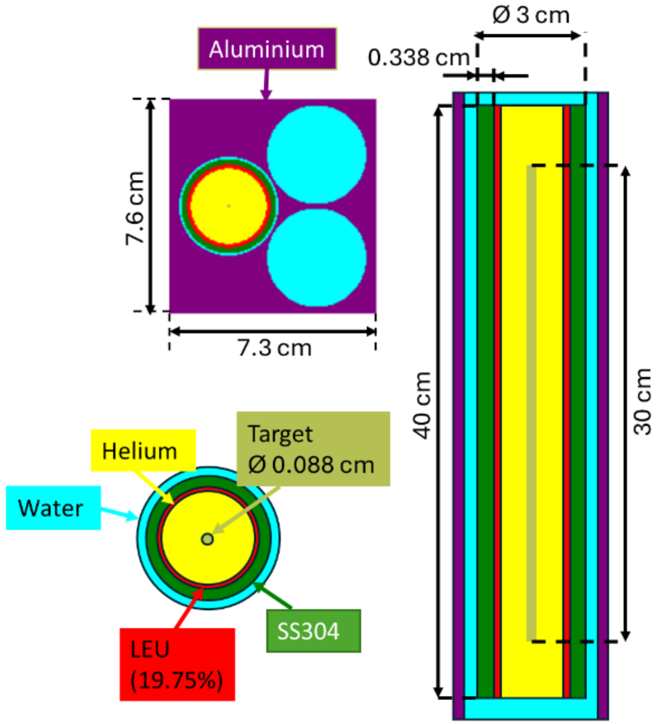
Fig. 3. First working core configuration of RSG-GAS.

Running Specifications

Fuel	19.45%U・U_3O_8-Al fuel plate
Thermal Power	15MWt
Neutron Flux	1.0E+13 [n/cm^2/s] (thermal neutron)

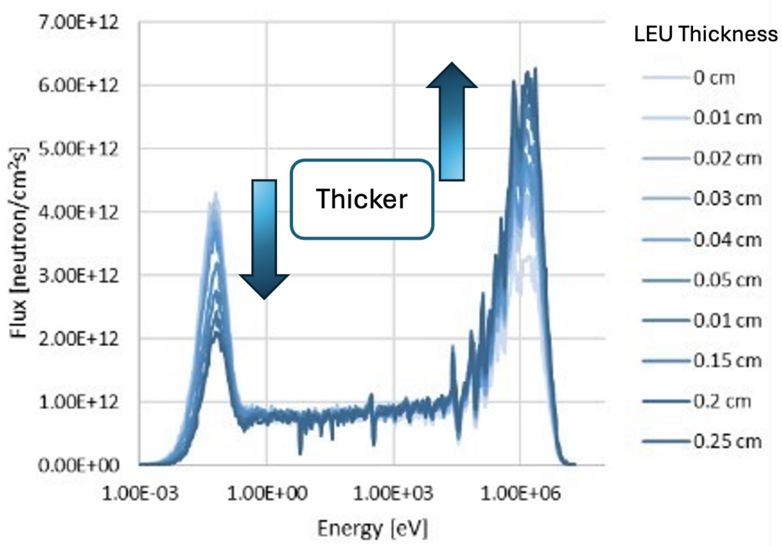
Code	OpenMC
Nuclear Data	ENDF/B VIII.0
Irradiation Time	26 days

Modified LEU Electroplating Capsule



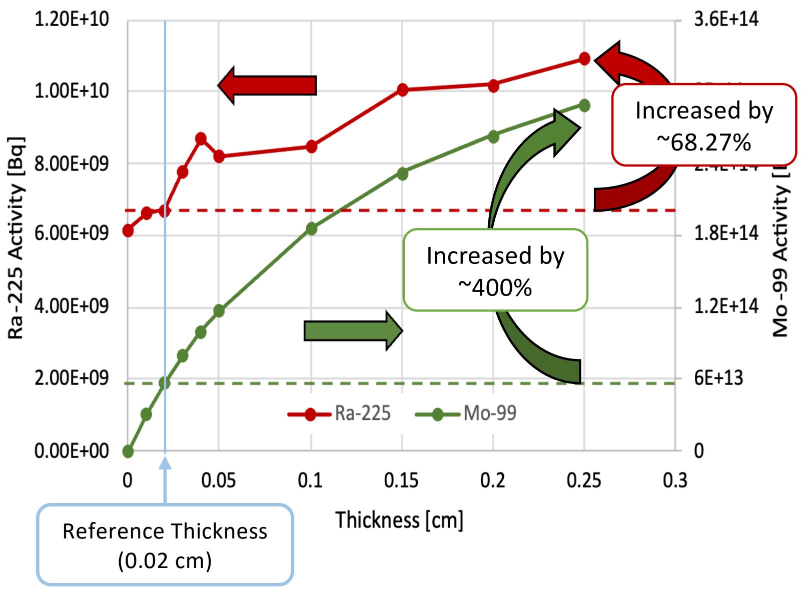
- Capsules with 0.02 cm LEU layer thickness are used for Mo-99 production from fission reaction.
- Simultaneous production of Ac-225 and Mo-99 by loading Ra-226 target into electroplating capsules is possible.
- In this research, the different thickness was considered, ranging from 0.01-0.25 cm.

Neutron Spectrum in Helium Gas



- This proved the main idea for this study, that the neutron spectrum was observed harder if the LEU layer was thicker, which was favorable for Ac-225 production.

Ra-225 and Mo-99 Production



Mo-99 = 6 days irradiation
Ra-225 = 26 days irradiation

- The method to change the LEU layer every 6 days should be taken into consideration for simultaneous production to occur.
- Ra-225 production at 0.25 cm increased by ~68.27% compared to 0.02 cm.
- Meanwhile, Mo-99 production at 0.25 cm was approximately 400% higher than 0.02 cm.

Conclusion

Existing fission reactors, Joyo and especially LWRs, have potentials to work as excellent facilities for producing medical isotopes, as by-products of electricity generation without needs for new facility construction and extra energy consumption.