

**Mo-99 2015 TOPICAL MEETING ON
MOLYBDENUM-99 TECHNOLOGICAL DEVELOPMENT**

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**Irradiation of LEU Targets in the BR2 Reactor
for Mo-99 Production**

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ABSTRACT

The BR2 reactor has currently with 7.800 six-day curies per week the largest installed irradiation capacity worldwide for the production of Mo-99. This production level is currently achieved by the irradiation of highly enriched uranium (HEU) targets in dedicated irradiation devices. In support of non-proliferation objectives and global reliability of supply, two separate projects have been initiated in 2012 with the processors IRE and MALLINCKRODT to develop low enriched uranium (LEU) targets for the production of Mo-99 without the use of HEU. The safety approval for 'test' irradiations has been received after submission and analysis of the safety report including neutronic calculations, thermohydraulic calculations, modification of existing irradiation devices, updated irradiation procedures, ... Five irradiation campaigns of LEU targets have been successfully performed in the BR2 reactor in 2014 before its refurbishment for a period of 16 months (February 2015 – June 2016) which will enable safe and reliable operation of the reactor for another period of at least 10 years. SCK•CEN is considering upgrading BR2's operating regime and increasing its yearly irradiation capacity for Mo-99 production if compatible with the full-cost recovery principle defined by OECD/NEA's High-Level Group on the Security of Supply of Medical Radioisotopes (HLG-MR).

1. Introduction

The BR2 reactor is a multipurpose 100 MW_{th} High-Flux 'Materials Testing Reactor' operated by the Belgian Nuclear Research Centre (SCK•CEN). The availability of high thermal neutron fluxes up to 10^{15} n·cm⁻²·s⁻¹ allows the production of a wide range of radioisotopes for various applications in nuclear medicine, industry and research such as Mo-99/Tc-99m, I-131, Xe-133, Lu-177, Sr-89, Re-186, Sm-153, Er-169, Y-90, W-188/Re-188, Sn-117m, P-32, Ir-192, Se-75, ... [1]

2. Current Mo-99 production

The BR2 reactor has currently with 7.800 six-day curies per week the largest installed irradiation capacity worldwide for the production of Mo-99 [2] as shown in Table 1.

Reactors	Countries	Targets	Weekly Irradiation Capacities [six-day curies]
BR2	Belgium	HEU	7.800
HFR	The Netherlands	HEU	4.680
NRU	Canada	HEU	4.680
SAFARI	South Africa	HEU/LEU	3.000
LVR-15	Czech Republic	HEU	2.800
OSIRIS	France	HEU	2.400
MARIA	Poland	HEU	2.200
OPAL	Australia	LEU	1.000
RA-3	Argentina	LEU	400

Table 1: Research reactors involved in the global Mo-99 production [2]

This production level is currently achieved by the irradiation of highly enriched uranium (HEU; 93% U-235) targets in dedicated irradiation devices according to an annual operating regime of 6 reactor cycles (20 operating weeks per year), i.e. 140 operating days per year. Six PRF (Primary Water Cooled Device for Reloadable Fissile Targets) dedicated irradiation devices are routinely loaded in reflector channels, providing a total irradiation capacity of 75 HEU targets per week. These devices are designed to allow loading/unloading of the 4 to 5 g U-235 HEU targets during reactor operation. A typical irradiation of 150 hours in a thermal neutron flux of $2.5 \cdot 10^{14} \text{ n}\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$ yields up to 1.000 Ci per target at "EOI" (End Of Irradiation), i.e. 120 six-day curies per target. After irradiation, the targets are moved from the 'irradiation position' to the 'cooling position' where they are cooled by the reactor primary water for 6 hours. They are then brought in the 'unloading position' and loaded into the transport containers at "EOI+12 hours". The containers are finally shipped to the processing facilities where Mo-99 ($T_{1/2}=66 \text{ h}$) and other fission radioisotopes as I-131 ($T_{1/2}=8.02 \text{ d}$) and Xe-133 ($T_{1/2}=5.24 \text{ d}$) can be recovered.

3. Refurbishment of the BR2 reactor

The BR2 reactor reached its first criticality in June 1961 and routine operation started in January 1963. The compact core is composed of a beryllium matrix providing 79 irradiation channels in which fuel elements, control rods, experimental and production devices are loaded. It is cooled and moderated by pressurized light water.

The BR2 reactor is currently temporarily shutdown for a period of 16 months (February 2015 – June 2016) for major refurbishment. The preventive replacement of the beryllium matrix will allow safe and reliable reactor operation for another period of at least 10 years.

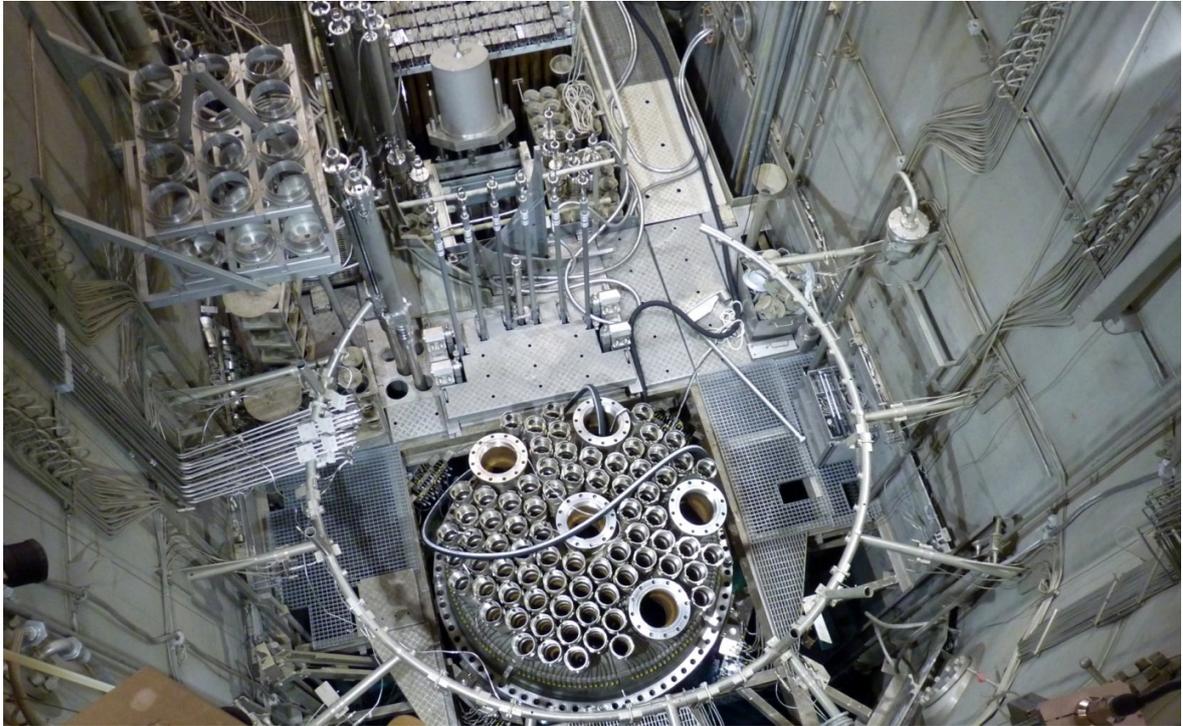


Figure 1: Unloading of the beryllium matrix from BR2's pressure vessel (April 2015)

Taking into account the upgrade that took place in the late 1960s, the current refurbishment is the fourth major maintenance of the BR2 reactor and the third replacement of the matrix.



Figure 2: New beryllium matrix installed in BR2's mock-up pressure vessel (August 2015)

The unloading of the fuel elements and the experimental devices from the reactor started mid-March 2015 (Figure 1). The first beryllium hexagon has been removed on the 1st of May 2015 from the pressure vessel and the last piece of beryllium end of June 2015. Meanwhile, the new beryllium matrix has been successfully installed in BR2's mock-up pressure vessel for dimensional checks in August 2015 (Figure 2). After all the required inspections – including the pressure vessel inspection programme – have taken place, the new beryllium matrix will be installed in BR2's pressure vessel begin of 2016. The programme is progressing very well according the established schedule which expects BR2 resuming routine operation from July 2016.

4. Future Mo-99 production

SCK•CEN is considering upgrading BR2's operating regime and increasing its yearly irradiation capacity for Mo-99 production if compatible with the full-cost recovery principle [3] defined by the OECD/NEA High-Level Group on the Security of Supply of Medical Radioisotopes (HLG-MR). An upgraded annual operating regime of up to 8 reactor cycles (27 operating weeks per year), i.e 190 operating days per year, would increase the potential production capacity (+ 35%) from 156.000 up to 210.600 six-day curies per year by the irradiation of HEU targets in a first phase and by the irradiation of LEU targets after their qualification expected in 2016-2017.

5. Test irradiations of LEU targets for Mo-99 production

In support of non-proliferation objectives and global reliability of supply, two separate and independent projects have been initiated in 2012 with the processors (MALLINCKRODT and IRE) and the target manufacturer (CERCA) to develop low enriched uranium (LEU; 19,75% U-235) targets for the production of Mo-99 without the use of HEU. The higher-density LEU targets specifications and geometries – confidential matter – have been determined in a consensus between the target manufacturer, the involved reactors and the processors taking all parameters into account as safety aspects, manufacturing constraints, impurities in the target material, irradiation positions, irradiation conditions, chemical processing, waste management, transportation issues, ... New irradiation baskets have been designed by SCK•CEN and manufactured to accommodate the LEU targets in BR2's existing PRF irradiation devices without too significant irradiation capacity loss (Figure 3).

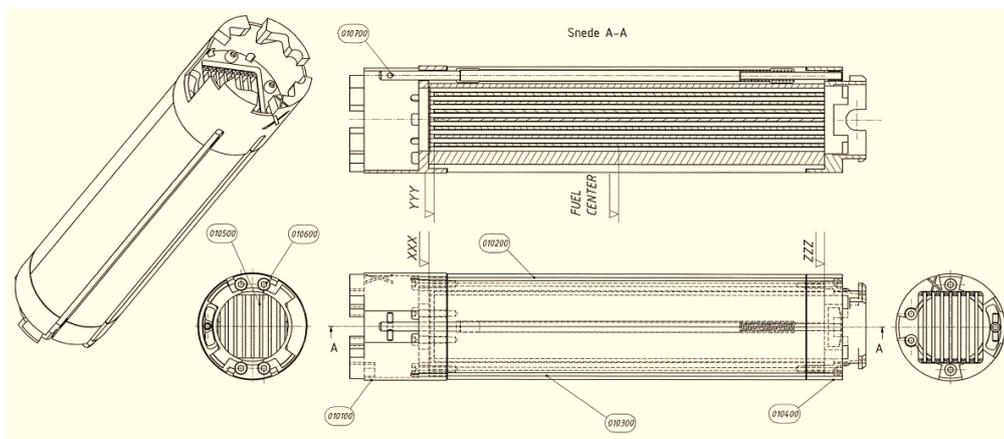


Figure 3: Irradiation basket for LEU targets

Detailed neutronic calculations have been performed by the MCNPX 2.7.0 [4] full core 3-D model of the BR2 reactor [5] to compare the isotopic vectors from the irradiation of HEU and LEU targets in representative conditions for future Mo-99 production (Figure 4).

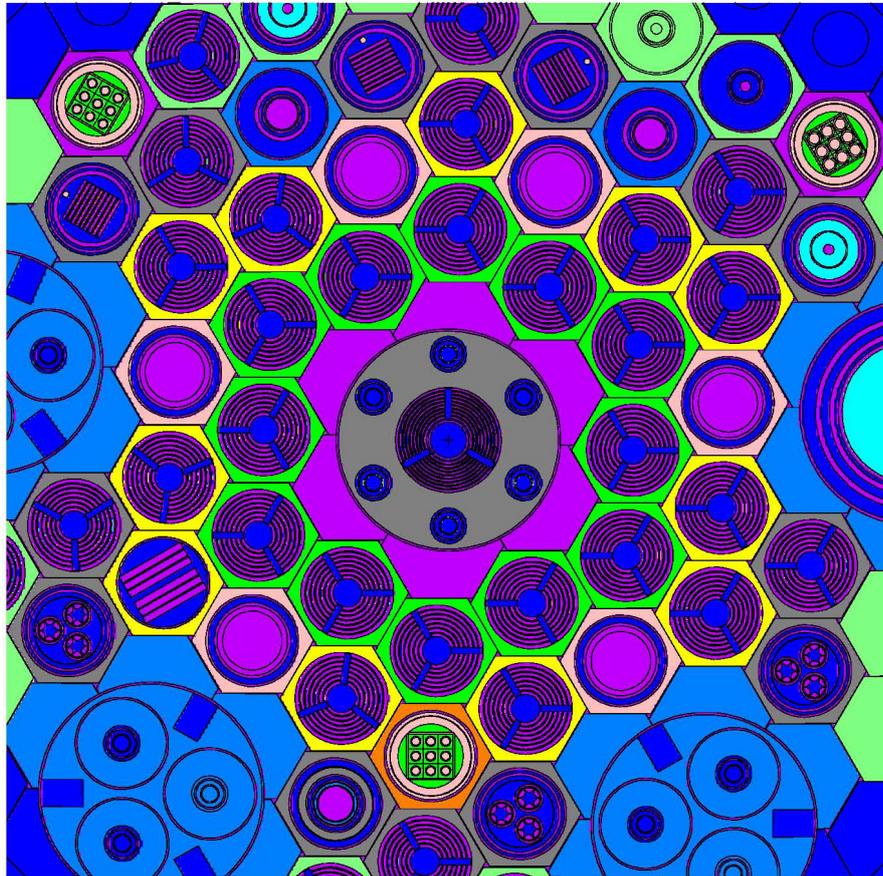


Figure 4: MCNPX 2.7.0 full core 3-D model of the BR2 reactor

The conversion of HEU into LEU targets would lead to decrease the Mo-99 activity produced per target by 20% to 25% at "EOI" (End Of Irradiation) and to increase the target consumption by the same factor to maintain an equivalent level of Mo-99 production. However, the six PRF irradiation devices will offer an increased weekly irradiation capacity of 78 LEU targets instead of 75 HEU targets currently. Hence, it is expected that the global loss of irradiation capacity for Mo-99 production in the BR2 reactor associated to the full conversion from HEU into LEU targets would be limited to approximately 15% to 20%.

Five test irradiation campaigns of LEU targets (34 targets in total) have been successfully performed in the BR2 reactor in 2014 before its refurbishment. The LEU targets have been irradiated for a duration of up to 200 hours. The maximum heat flux on the targets has been increased gradually to reach a level close to the current safety limit fixed by the Safety Committee for the irradiation of targets in a PRF device.

The analysis of water samples taken during the unloading of the LEU targets demonstrated that there was no fission products release. The visual inspection showed that there was no issue related to the irradiation and handling procedures (Figure 5).

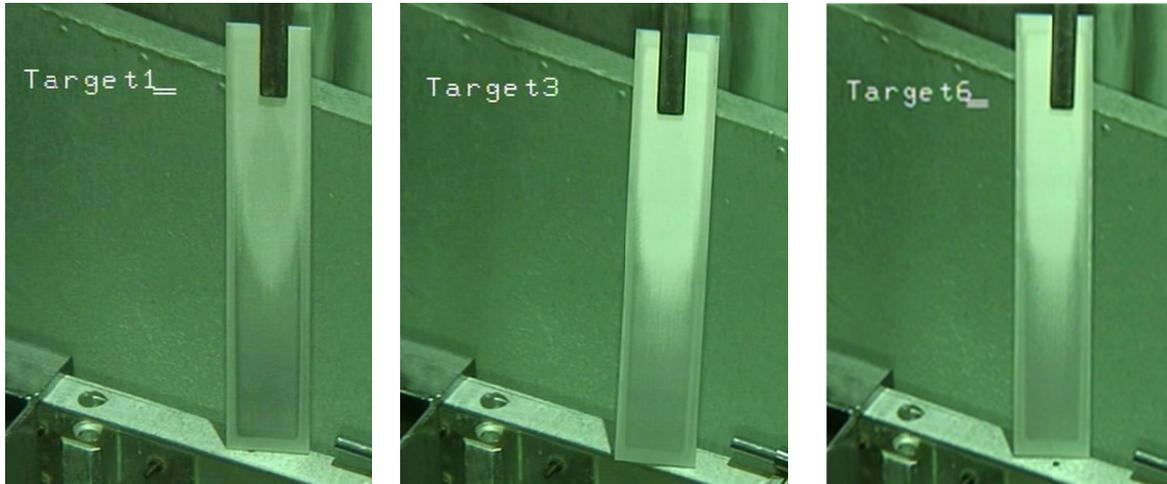


Figure 5: Visual inspection of irradiated LEU targets

The non-destructive "PIE" (Post Irradiation Examination) showed no swelling and only minor oxide growth (Figure 6).

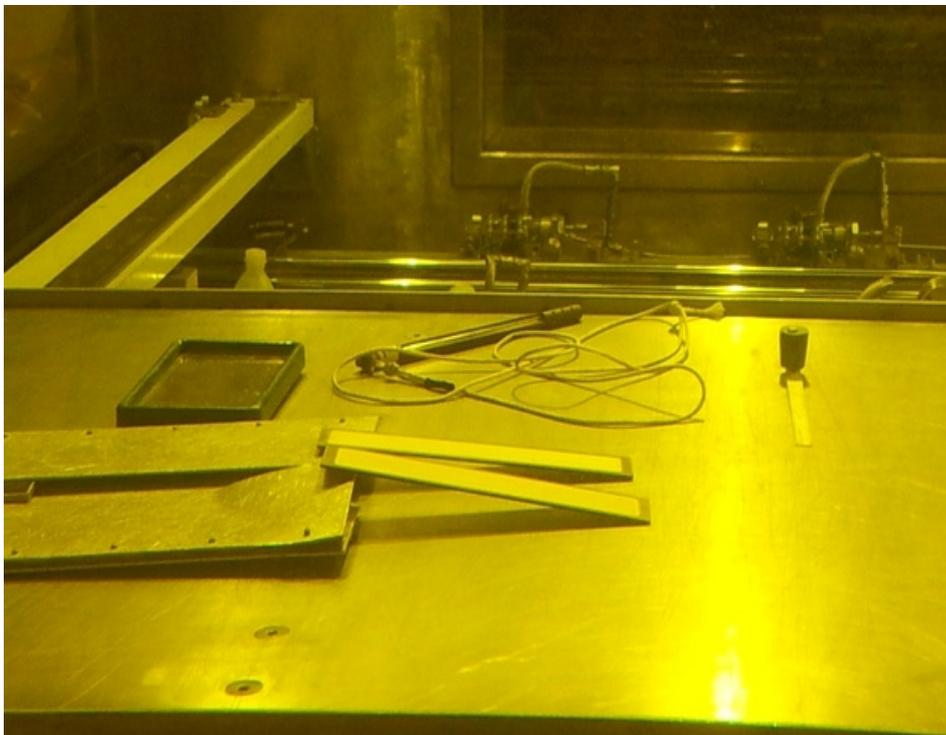


Figure 6: Non-destructive "PIE" of irradiated LEU targets

6. Conclusion

Five irradiation campaigns have been successfully performed in the BR2 reactor in 2014 for the qualification of LEU targets. The full conversion from HEU into LEU targets for Mo-99 production will be gradually achieved in agreement with the processors MALLINCKRODT and IRE in the period 2016-2017. The loss of irradiation capacity for Mo-99 production in the BR2 reactor would be limited to approximately 15% to 20% by the conversion process. However, the refurbishment of the BR2 reactor currently performed during 16 months in 2015-2016 will contribute substantially to the long-term and reliable supply of Mo-99 until 2026 at least.

7. References

- [1] B. Ponsard, "Production of Radioisotopes in the BR2 High-Flux Reactor for applications in Nuclear Medicine and Industry", 9th IIS International Symposium on the Synthesis and Applications of Isotopes and Isotopically Labelled Compounds, Edinburgh, Scotland, July 16-20 2006, Journal of Labelled Compounds and Radiopharmaceuticals, Volume 50, Issue 5-6, pp. 333-337, ISSN 0362-4803, April-May 2007.
- [2] NEA/SEN/HLGMR (2014) 2, "The Supply of Medical Radioisotopes – Medical Isotope Supply in the Future: Production Capacity and Demand Forecast for the Mo-99/Tc-99m Market, 2015-2020", OECD/NEA, www.oecd-nea.org, April 2014.
- [3] NEA/SEN/HLGMR (2012) 9, "Full-Cost Recovery for Molybdenum-99 Irradiation Services : Methodology and Implementation", OECD/NEA, www.oecd-nea.org, February 2012.
- [4] "MCNPX Version 2.7.0, LANL, LA-UR-11-02295. April, 2011.
- [5] S. Kalcheva, E. Koonen and B. Ponsard, "Accuracy of Monte Carlo Criticality Calculations During BR2 Operation," Nuclear Technology, 151 (2005), p. 201-219, ISSN 0029-5450.