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

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MARIA Research Reactor in Supply Chain of Mo-99

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ABSTRACT

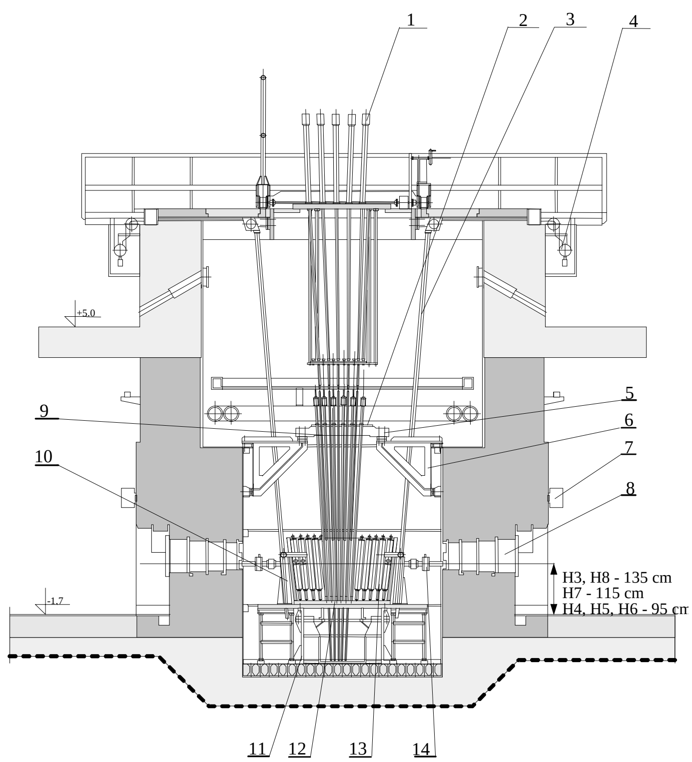
The high flux research reactor MARIA is operated at the National Centre for Nuclear Research. Due to supply shortages of molybdenum-99 during July 2009 – February 2010 there was developed the Mo-99 irradiation and transport technology in MARIA reactor facility and then its expedition to the processing facility in Petten (The Nederlands). From February 2010 till the end of 2014 were irradiated 1112 uranium plates. Especially important was MARIA reactor operation from the mid of December 2012 till the end of April 2013 when the quantity of delivered for market Mo-99 covered about 15% of global demand. After upgrading of the primary fuel channel cooling system we are available to install an additional channel for U-targets irradiation (increased capacity around 50%). Under collaboration with Mallinckrodt Pharmaceuticals and with reactors HFR and BR-2 we are developing irradiation and transport technology of a new designed LEU targets for molybdenum production.

1. Introduction

Due to the shutdown of the NRU reactor (Canada) and plans for scramming the HFR reactor (The Nederlands) in the half of 2009 a decision was taken on cooperation between IAE and COVIDIEN which was aimed to initiate an irradiation of high-enriched uranium plates in MARIA reactor for production of molybdenum Mo-99. There was developed the Mo-99 irradiation and transport technology in MARIA reactor facility and then its expedition to the processing facility in Petten. The physics calculation, safety analyses, technical design of equipment for irradiation and transport inside the reactor facility and loading to the special transport container (MARIANNE) were made.
We received the positive opinion of the Nuclear Safety Committee of IAE and approvals released by the National Atomic Energy Agency in Poland.
The first irradiation of uranium plates has been taken place in the MARIA reactor from February 2010. In 2010 in MARIA reactor we have made 21 cycles of U-targets irradiation for Mo production. Under supervision of the Mallinckrodt Pharmaceuticals we are developing irradiation and transport technology of a new LEU targets.

2. MARIA research reactor description

The research reactor MARIA is operated at the National Centre for Nuclear Research. The multipurpose high flux research reactor MARIA is a water and beryllium moderated reactor of a pool type with graphite reflector and pressurised channels containing concentric tubes assemblies of fuel elements. It has been designed to provide high degree of flexibility. The fuel channels are situated in a matrix containing beryllium blocks and enclosed by lateral reflector made of graphite blocks in aluminium cans, *FIG.1*. The MARIA reactor is equipped with vertical channels for irradiation of target materials, a rabbit system for short irradiations and six horizontal neutron beam channels.



1. control rod drive mechanism
2. mounting plate
3. ionization chamber channel
4. ionization chamber drive mechanism
5. fuel and loop channels support plate
6. plate support console
7. horizontal beam tube shutter drive mechanism
8. beam tube shutter
9. fuel channel
10. ionization chambers shield
11. core and support structure
12. core and reflector support plate
13. reflector blocks
14. beam tube compensator joint

FIG. 1. Vertical cross-section of MARIA RR.

The main characteristics and data of MARIA reactor are as follows:

- nominal power — 30 MW (th),
  thermal neutron flux density — $3.0 \cdot 10^{14}$ n / cm^2 · s,
- moderator — H_2O, beryllium,
- cooling system — channel type,
- fuel assemblies:

- material U_3Si_2
- enrichment 19,75 %
- cladding aluminium alloy
- shape five concentric tubes
- active length 1000 mm

▪ output thermal neutron flux
at horizontal channels $3 \div 5 \cdot 10^9$ n / cm^2 · s

The reactor was fully converted from HEU to LEU fuel in the end of August 2014.

In the end of March 2015 reactor MARIA received the new license for reactor operation till the end of March 2025.

The main areas of reactor application are as follows:

- production of radioisotopes,
- research in neutron and condensed matter physics,
- neutron radiography,
- neutron activation analysis,
- neutron transmutation doping,
- testing of fuel and structural materials for nuclear power engineering.

3. Developing the uranium targets irradiation technology, calculations, safety analysis, measurements and tests

In the period from June 2009 up to January 2010 the technology and safety analysis of irradiation and shipment of uranium was developed and also a number of tests and measurements were conducted.

Calculations and safety analyses at steady states were as follows:

- safety analysis and design of irradiation rig,
- safety analysis of transport of plates into MARIA hot cell to processing facility using special transportation container,
- calculations of molybdenum activity,
- neutronic calculations,
- thermal-hydraulic calculations at steady states,
- activity of fission products and thermal power of the uranium plate batch,
- cooling of uranium plates in the capsule for irradiation during natural convection in the air,
- shielding calculations and an assessment of radiological hazard for personnel pending reloading – transport operations.

Program of examinations and installation tests consisted of:

- hydraulic measurement of channel for irradiation of containing the mock-ups of plates,
- cold trials of reloading and transport operations with a batch of dummy plates,
- calibration measurements of calorimeter for measuring of thermal power of 4 plate batch,
- measurement of the heat balance in molybdenum installation with the dummy plates,
- test irradiation of uranium plates and their dispatching,
- measurements of temperatures of uranium plates in the air.
- handling and loading of plates into the transport Marianne cask,
- engineering of irradiation facility & reloading devices,

Testing irradiation were made in the beginning of February 2010 and in the near future the U.S. Food & Drug Administration and Health Canada have approved the use MARIA RR as a site of irradiate for ^{99}Mo.

4. Current status of Mo-99 production in the MARIA reactor

In the period March 2010 to the end of 2014, 76 irradiation cycles in the MARIA reactor were conducted. In all cycles were irradiated 1112 plates.

Irradiations were conducted in two different locations of molybdenum channels (f-7 and i-6) and different configurations of the core. The typical configuration of the core for irradiations of molybdenum channels is shown in Fig.2.

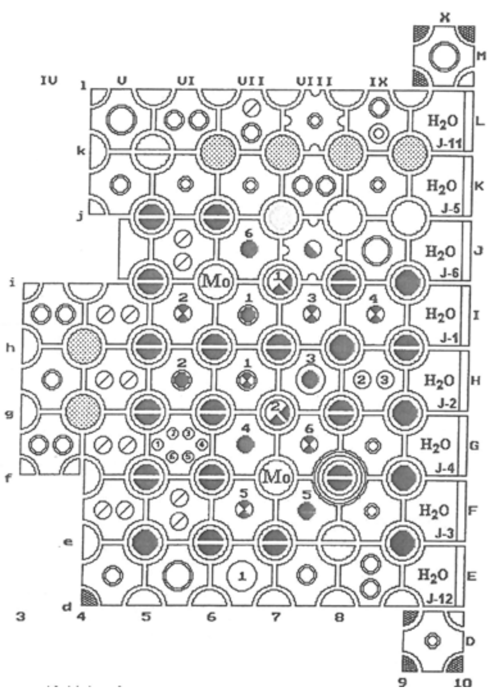


FIG. 2. A typical configuration of the MARIA reactor core with molybdenum channels.

The details of irradiation cycles of uranium plates (8pcs) in the Maria reactor are presented below:

- time of irradiation – 120 [hours]
- average power generated – 180 ÷ 200 [kW]
- Mo-99 activity at the end of irradiation (EOI) – 7000 ÷ 8000 [Ci]

The typical technological parameters of MARIA RR core are shown in *FIG.3*.

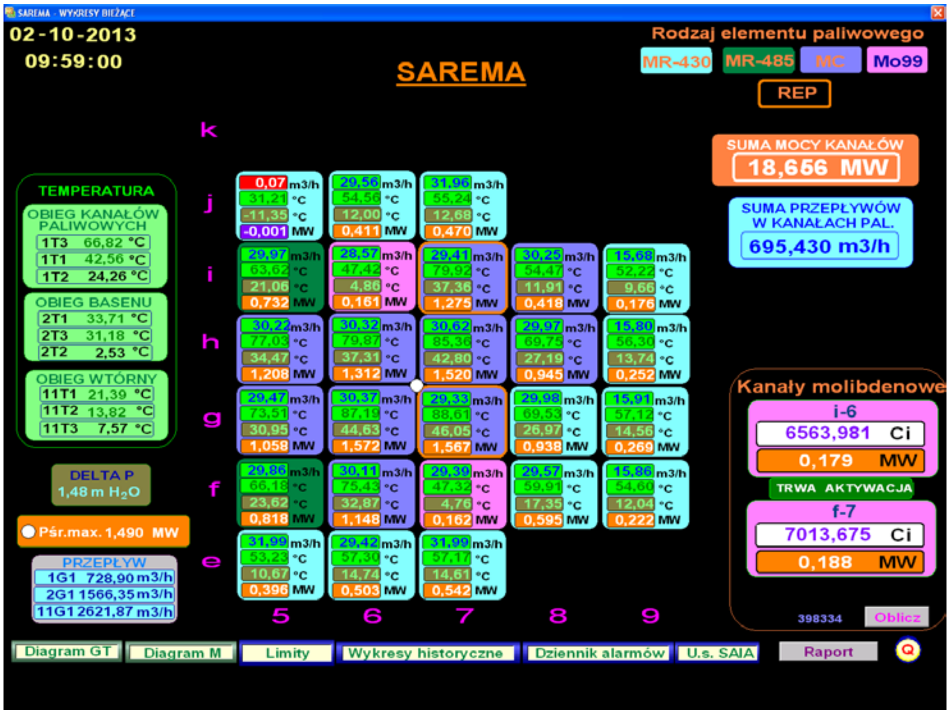


FIG. 3. Diagram of MARIA RR core.

The programme of HEU targets irradiations in 2015 connected with 20 cycles is shown in *FIG.4*.

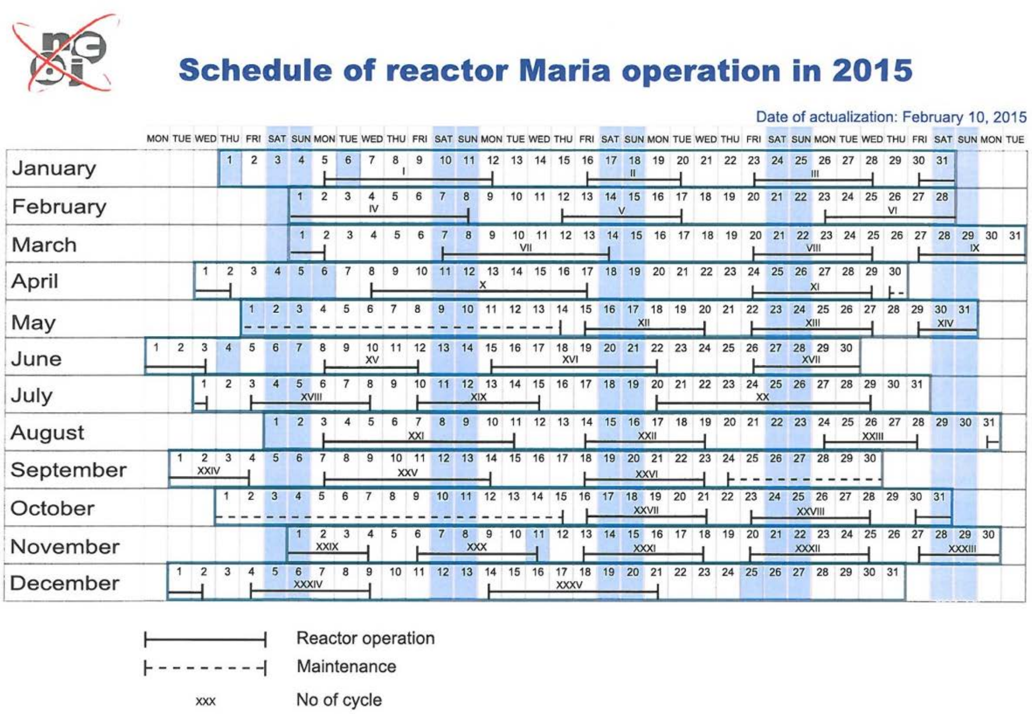


FIG. 4. Schedule of reactor MARIA operation in 2015.

In the beginning of July 2015 we increased Mo-99 production capacity by increasing the number o containers with uranium plates in the channel (from 8 to 12 pcs).
Average power was increased to 240 kW and Mo-99 activity in the EOI was increased to 9200 Ci.

5. Irradiation of HEU IRE targets in MARIA

We have started cooperation between NCBJ and IRE, Belgium to develop the technology of U-targets irradiation, handling and loading to the AGNES transport containers and then expedition of irradiated targets to the processing facility in Flerus (Belgium).
The scope of program includes:

- safety analysis (neutronic, thermohydraulic in steady states, transient and emergency situation),
- technology of irradiation handling and reloading of irradiated targets:
  - manufacturing and commissioning of an irradiation facility,
  - licensing process (interaction between NCBJ and Regulators Authority
  - training of MARIA reactor operators,
  - hot tests.

We plan to irradiate HEU IRE targets tubes in position h-8 of MARIA reactor core, FIG. 2.

6. Qualification of the new LEU targets.

Under supervision of the Mallinckrodt Pharmaceuticals and with collaboration with HFR and BR2 reactors we are developing irradiation and transport technology of a new designed Low Enriched Uranium (LEU) targets for molybdenum production.
The first step of works comprised of development of technology and neutronic and thermal-hydraulic calculations.:
Technology over irradiation, reloading and transport of LEU plates comprise of:

- irradiation of plates and preliminary cooling in the channel,
- calorimeter measurement of the residual power generation,
- transport of plates to the hot cell,
- measurement uranium plates temperature,
- reloading handling operation in the hot cell,
- emergency procedure in case of damaging the uranium plates in the cell,
- loading the plates into the container MARIANNE.

Neutronic and thermal-hydraulic calculations at the steady-states and analysis of transients consist of:

- reactor kinetic parameters: λ_{eff} and β_{eff}
- hydraulic characteristics,
- forced coolant flow
- natural convection,
- cooling of low enrich uranium LEU in the can for irradtion under natural convection in the air,
- an assessment of radiological hazard during transport handling operation
- influence of the reactor transient states linked with insertion of positive reactivity on irradiation conditions over LEU plates,
- lose of leek tightness of the LEU plate during irradiation in the channel,

- analysis of abnormal states during discharging and transport of cans from the channel for irradiation into the hot cell,
- analysis of abnormal states during handling operations in the hot cell,
- analysis of radiological hazard associated with damaging LEU plates in the hot cell,
- limits revelant to LEU uranium plates,
- thermal hydraulic limits over uranium HEU and LEU plates' irradiation,
- thermal limits during execution of reloading and transport operations,
- constraints and limits associated with radiological hazard during the relocating and transport operations.

The second step will be modification of the design existing molybdenum channel and manufacturing the new parts of the irradiation rig.
The third step will be licensing process (interactions with the Regulatory Authority)
The fourth step will be:

- upgrading of expedition devices including modification inner transportation container
- modification of existing equipment in the Hot Cell
- adaptation of the hoisting device for MARIANNE container with new inner container

The final step will be test irradiation of 8 LEU plates and its expedition to Petten facility.

6. Conclusion

From the 2010 when we started irradiation of HEU U-targets MARIA research reactor is in the supply chain to ensure a reliable supply of molybdenum-99 for nuclear medicine.
After upgrading of the pumps in the primary cooling system we increased capacity of Mo-99 irradiation in MARIA reactor around 50%.
In the near future we will start to irradiate HEU targets for the new partner.
The specific activity of U-targets reached in MARIA is very high and the period December 2012 – April 2013, we delivered for market Mo-99 covered about 15% of global demand.
We are working on qualification of LEU target irradiation and transport technology in MARIA reactor and than its expedition to the processing facility in Petten (The Nederlands).