IRRADIATIONS OF HEU TARGETS IN MARIA RR FOR MO-99 PRODUCTION

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1. INTRODUCTION

Due to the shutdown of the NRU reactor in Canada and plans for shutting down the HFR reactor in the Netherlands in the latter 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 reprocessing factory in Petten, Netherlands. The physics calculation, safety analyses, technical projects of equipment for irradiation and transport inside the reactor facility and loading to the special transport container (MARIANNE) were made.

After receiving the positive opinion of the Nuclear Safety Committee of IAE and approvals released by the National Atomic Energy Agency there were made:

- Channel for uranium plates’ irradiation;
- Equipment to be used for plates’ transport from the reactor core to the hot cell;
- Reloading stand for the transport container, MARIANNE; and
- Rail trolley for transport and lifting of shielding container MARIANNE.

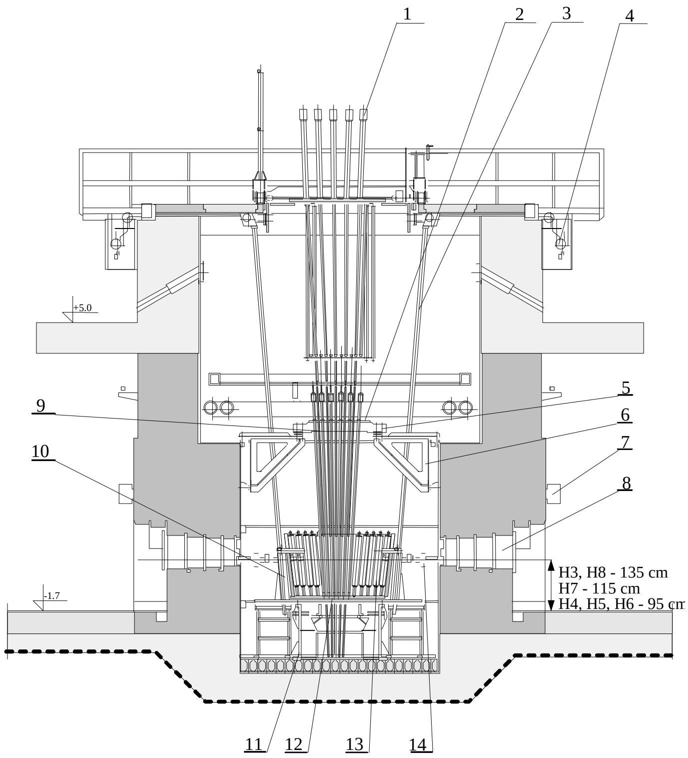
It was accomplished the program for checking and testing full installation which included:

- Loading unloading and transport of plates within the boundary of reactor pools;
- Reloading of the plate dummies to the shielding container;
- Irradiation of the plates’ dummies (Al) in the reactor; and
- Monitoring of container leak-tightness.

The number of licenses needed for the transport of irradiated uranium plates from MARIA reactor in Poland through Germany to Holland was received.

2. MARIA RESEARCH REACTOR DESCRIPTION

The research reactor MARIA is operated at the Institute of Atomic Energy POLATOM (IAE). 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 six-tube 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, Figure 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: 4.0×10^{14} $cm^{-2}s^{-1}$;
- Moderator: H_2O, beryllium;
- Cooling system: channel type;
- Fuel assemblies:
  - Material: UO_2-Al alloy;
  - Enrichment: 36%;
  - Cladding: Aluminium;
  - Shape: Six concentric tubes;
  - Active length: 1000 mm;
- Output thermal neutron flux:
  - At horizontal channels: 3.5×10^{9} $cm^{-2}s^{-1}$.

The main areas of reactor application are as follows:

- Production of radioisotopes;
- Testing of fuel and structural materials for nuclear power engineering;
- Neutron radiography;
- Neutron activation analysis;
- Neutron transmutation doping;
- Research in neutron and condensed matter physics.

3. DEVELOPING THE URANIUM TARGETS IRRADIATION TECHNOLOGY, SAFETY ANALYSIS, MEASUREMENTS AND TESTS

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

Technology for irradiation and handling of uranium plates comprise of:

- Irradiation of plates and initial cooling in the irradiation channel;
- Calorimetric measurement of heat generation in the capsule with plates;
- Transport of plates into the hot cell;
- Handling operations in the hot cell; and
- Loading of plates into the transport cask MARIANNE.

Calculations and safety analyses at steady states are as follows:

- 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; and
- Shielding calculations and an assessment of radiological hazard for personnel pending reloading and transport operations.

Program of examinations and installation tests consist of:

- Hydraulic measurement of channel for irradiation of capsules containing the mock-ups of plates;
- Cold trials of reloading and transport operations with a bath of dummy plates;
- Calibration measurements of calorimeter for measuring of thermal power of 4 plate batch;
- Measurement of axial distributions of the neutron flux density in the capsule containing dummy plates;
- Measurements of the heat balance in molybdenum installation with the dummy plates;
- Test irradiation of uranium plates and their dispatching; and
- Measurements of temperatures of uranium plates in the air.

The following assumptions have been taken into account while developing the uranium targets irradiation technology for the production of the molybdenum in the MARIA reactor:

(i) Irradiation is held in containers, containing 4 plates each, loaded into the molybdenum channel. Currently (June 2010) in the MARIA reactor there are two molybdenum channels, intended to irradiate 2 containers each.

(ii) Irradiation of the containers with plates is held in installations which are converted fuel channels of the MARIA reactor. Thermal power generated in 8 uranium plates loaded into 2 containers, is ~ 220 kW.

(iii) Adaptation of the MARIA reactor fuel channel for the purpose of irradiation of uranium plates consists in such an alteration of the channel so that it is possible to repeat loading and discharge of the containers with plates from the installation without the necessity of the evacuation of the irradiation channel from the reactor core.

(iv) The nominal flow of coolant is maintained in the irradiation installation because of high thermal flux and the possibility of the molybdenum channel location change.

(v) Measurements of temperatures and the coolant flow in the molybdenum installation carried out in the centralized SAREMA control system don't give the direct information about the current fission power generated in uranium plates because they also take into account heat generation from the gamma radiation from neighboring fuel elements as well as heat exchange between channels and the reactor pool.

(vi) Preliminary cooling of uranium plates after irradiation is held in the installation to irradiation as part of the procedure of removing of the residual heat generated in fuel of the MARIA reactor. The cooling is ensured by the circuit of cooling fuel channels. Opening of the molybdenum channel and the evacuation of the containers with uranium plates are held of not earlier than 10 hours after the reactor shutdown.

(vii) The transport of the containers with plates into the hot cell involves the change of conditions of cooling the containers with plates as well as uranium plates. Handling operations in the hot cell are conducted in the air. Cooling plates with the natural convection in the air is less efficient than cooling convection in water. The recipient of uranium plates determines two thermal limits for the set of 8 uranium plates: residual power 548 W, (it is possible to start the transport of plates in the air and loading into the transport container MARIANNE) and residual power 450 W (transport of the container MARIANNE along highway roads is allowed). The procedure of the uranium plates dispatch includes the possibility of conducting calorimetric measurements of the residual heat generated in a single container with plates.

(viii) On account of the uncertainty resulting from heat exchange in conditions of the natural convection in the air the measurements of plate's temperature were conducted in hot cell during test irradiation. These measurements showed that temperatures of uranium plates in the air were below 200°C.

(ix) The total activity of fission products in uranium plates during transport operations in the hot cell is approximately 100 kCi. Test measurements showed that the shielding of the hot cell is sufficient for safe performing handling operation of irradiated uranium plates.

(x) After the process of irradiation and cooling plates has ended the plates are loaded into a special shielding container MARIANNE and transported to COVIDIEN company for reprocessing at the laboratory in Petten (Netherlands).

4. MO-99 PRODUCTION IN THE MARIA REACTOR — CURRENT STATE

In the period from 11 March–2 June, 2010, 8 irradiation cycles in molybdenum channels in the MARIA reactor were conducted. In all cycles 12 sets of uranium plates (8 plates in each) were irradiated. During 8–14 February, 2011, the test irradiation of 8 plates was conducted.

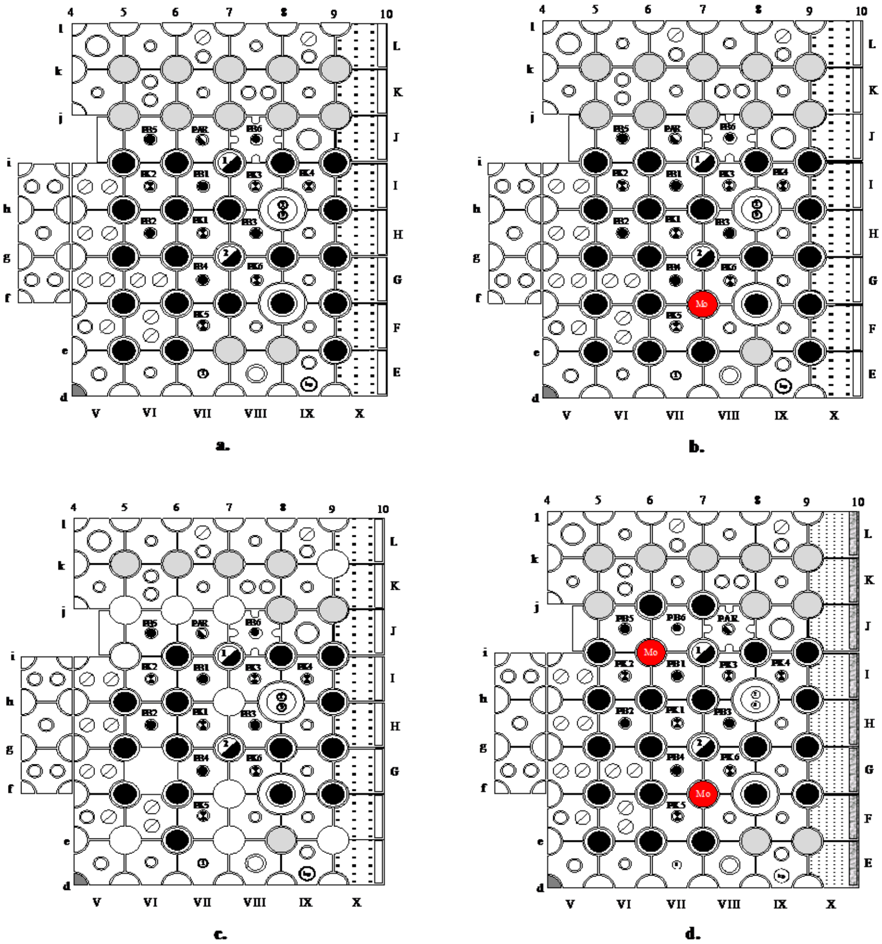


Fig. 2. Configurations of the MARIA reactor core with molybdenum channels.

Irradiations were conducted in three different locations of molybdenum channels (f-7, h-7 and i-6) and different configurations of the core, introduced in *FIG.2*. The configuration of the core preceding cycles of the work with molybdenum channels also shown in the picture (Figure 2a.).

The details of irradiation cycles of uranium plates in the Maria reactor (along with test irradiation) are presented below:

- Time of irradiation: 135–145 h;
- Average power: 180–200 kW; and
- Mo-99 activity at the end of irradiation (EOI): 7500–8000 Ci.

The technological parameters of MARIA RR core are shown in Figure 3.

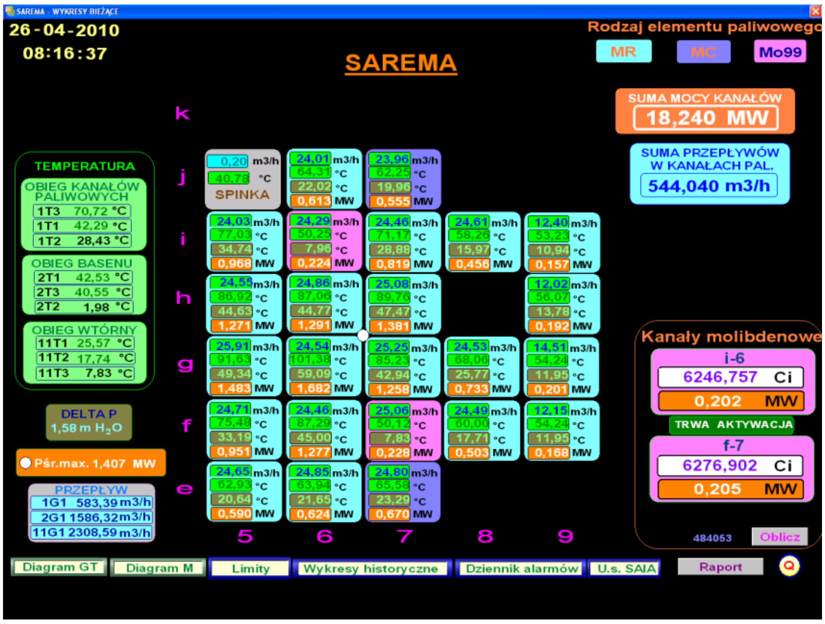


Fig. 3. Diagram of MARIA RR core.

The programme of HEU targets irradiations in 2010 connected with 20 cycles is shown in Figure 4.

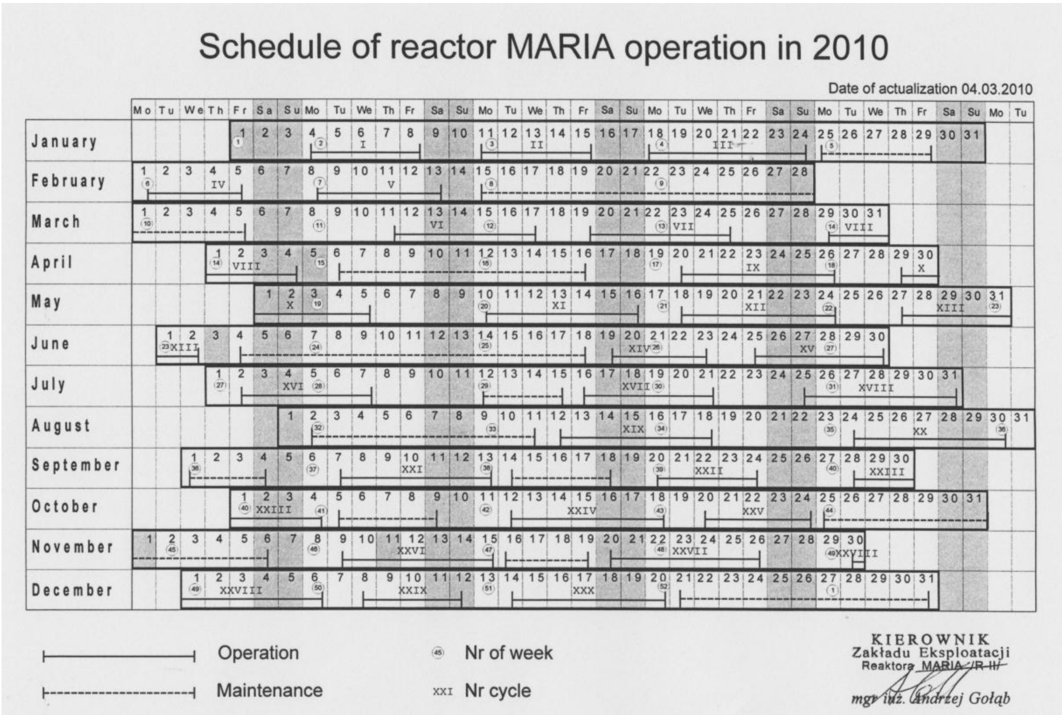


Fig. 4. Schedule of reactor MARIA operation in 2010.

The installation of molybdenum channels in the MARIA reactor core involving replacement of fuel channels with molybdenum channels causes decrease of excess reactivity of the core. This fall can be compensated either by the partial exchange of fuel for less burnt-out or increasing the number of fuel elements. In the two first irradiation cycles (2010/V and 2010/VI) the partial exchange of fuel for less burnt-out was made. Installation of two molybdenum channels (cycles 2010/VII and 2010/VIII) required loading into the core two additional fuel channels. All these changes involve the increasing of mass of U-235 in such a way to provide the appropriate excess reactivity of the core.

After reactor shut down and cooling time minimum 12 hours the irradiation holders are unloading from the channels and handling to the dismantling cell.

Calorimetric measurements conducted directly before the dispatch of plates showed residual powers in the range of 320-410 W, that is below limit 450 W of power, which makes it possible to transport 8 irradiated uranium plates in the container MARIANNE.

Measurements of the temperature of plates in the container in conditions of the natural convection in the air were made after test irradiation has ended. The measurement was performed directly before plates were loaded into the shielding container MARIANNE. The temperature of plates didn't achieve the value of 200°C.

Then the irradiated targets are loaded to the Marianne container (Figure 5).

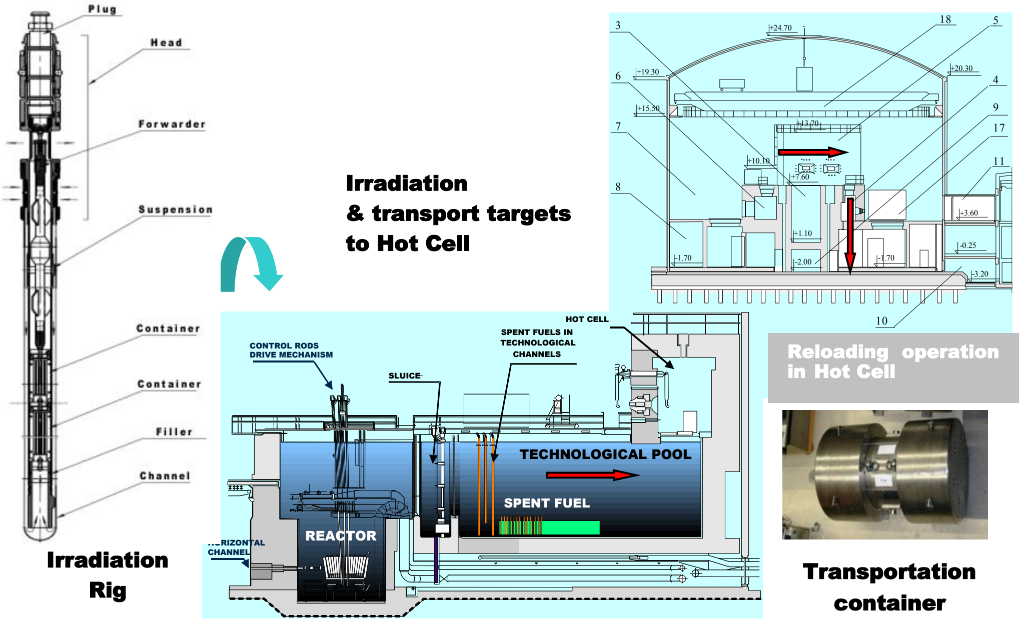


Fig. 5. Handling of irradiated target in MARIA reactor building.

After vacuum and leak tests the Marianne container is ready for transport truck from Świerk site to Petten.

5. CONCLUSION

The realization of the molybdenum program confirmed the correctness of the irradiation technology and handling operations in the reactor pools and in hot cell as well as the loading operation into the transport container MARIANNE. Experience acquired made it possible to implement additional technical and organizational solutions, which increased the certainty and shortened the time of handling operations not violating adopted operating procedures.

The achieved very good results of production are an important step in increasing commercial products and services of MARIA research reactor.