

Institute of Atomic Energy POLATOM OTWOCK-SWIERK POLAND

Irradiations of HEU targets in MARIA RR

for Mo-99 production

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INTRODUCTION

- In May 2009 the NRU reactor (Canada) was shutdown and was planned for scramming the HFR reactor (Holland),
- In the half of 2009 a decision was taken on cooperating between IAE and COVIDIEN,
- IAE and COVIDIEN initiative cover an irradiation of highenriched uranium plates in MARIA reactor for production of molybdenum Mo-99 in Petten,
- There was developed the Mo-99 irradiation and transport technology in MARIA reactor facility and then its expedition to the reprocessing factory in Petten (Holland).



INTRODUCTION



Irradiation facility manufacture



MARIA RESEARCH REACTOR Facility description

- The high flux rector MARIA is a water and beryllium moderated reactor of 30 MW power level;
- Pool type reactor with pressurized fuel channels containing concentric tube assemblies of fuel elements;
- Fuel channels are situated in matrix containing beryllium blocks surrounded by graphite reflector;
- Main characteristics and data of MARIA reactor:
 - maximum power
 - thermal neutron flux density
 - moderator
 - reflector
 - cooling system

30 MW (th) $2,5 \times 10^{14} \text{ n/cm}^2 \text{ s}$ H₂O, beryllium graphite in Al channel type





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
- core and support structure
 core and reflector support plate
- 13. reflector blocks
- 14. beam tube compensator joint

FIG. 1. Vertical section of the MARIA reactor.



The main areas of reactor application are as follows:

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



80% / 36% U-235 (from 1999)

FUEL ELEMENT - MR

- Material UO_2 -Al. Alloy clad in aluminium
- Enrichment
- Shape concentric tubes
- Dimensions 1000 mm height / 79 mm diameter
- Cooling under pressure flow
- Power limited to 1.8 MW
- 2 Lead Test Assemblies MC-LEU (19,7% U-235) are irradiated for qualification





FIG. 2. MR - Fuel element



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
- 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
- Shielding calculations and an assessment of radiological hazard for personnel pending reloading – 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
- Measurements of temperatures of uranium plates in the air



- Irradiation is held in containers, containing 4 plates each, loaded into the molybdenum channel.
- Irradiation of the containers is held in installations which are converted fuel channels of the MARIA reactor.
- Loading and discharge of the containers with plates from the installation is possible without the necessity of the evacuation of the irradiation channel from the reactor core.
- The nominal flow of coolant is maintained in the irradiation installation.



- 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.
- 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 and 450 W).
- 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.
- 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.



- The total activity of fission products in uranium plates during transport operations in the hot cell is ca. 100 kCi.
- Test measurements showed that the shielding of the hot cell is sufficient for safe performing handling operation of irradiated uranium plates.
- After the process of irradiation and cooling plates has ended the plates are loaded into a special shielding container MARIANNE



MO-99 PRODUCTION IN THE MARIA REACTOR – CURRENT STATE

- Between 8th ÷ 14th February the test irradiation of 8 plates has been conducted.
- In the period from 11th March to 2nd June 2010 eight 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.
- 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.3*.





FIG. 3. Configurations of the MARIA reactor core with molybdenum channels.



MO-99 PRODUCTION IN THE MARIA REACTOR – CURRENT STATE

- The details of irradiation cycles of uranium plates in the Maria reactor (along with test irradiation) are presented below:
 - time of irradiation 135÷ 145 [hours]
 - average power

- 180 ÷ 200 [kW]
- Mo-99 activity at the
 end of irradiation (EOI) 7500 ÷ 8000 [Ci]



Schedule of reactor MARIA operation in 2010

Date of actualization 04.03.2010

	Mo	Τu	We	Τh	Fr	Sa	Su	Мо	Tu	We	Th	Fr	Sa	Su	Мо	Tu	We	Th	Fr	Sa	Su	Мо	Tu	We	Th	Fr	Sa	Su	Мо	Tu	We	Th	Fr	Sa	Su	Мо	Tu
January					1	2	3	4	5	6 I	7	8	9	10	11	12	13 11	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	31		
February	1	2	3	4 IV	5	6	7	8 (7)	9	10	11 V	12	13	14	15 	16	17	18	19	20	21	22	23	24	25	26	27	28							144		
March	1	2	3	4	5	6	7	8	9	10	11	12	13 VI	14	15	16	17	18	19	20	21	22 (13	23 VI:	24	25	26	27	28	29	30 VI	31 II	1					
April				1	2 VII	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19 (17)	20	21	22	23 IX	24	25	26	27	28	29	30 X	1			
May						1	2 X	3 (19)	4	5	6	7	8	9	10	11	12	13 XI	14	15	16	17	18	19	20	21 XII	22	23	24	25	26	27	28	29 XII	30	3 1 (23)	
June		1 (23)X	2	3	4	5	6	7 (24)	8	9	10	11	12	13	14	15	16	17	18	19 -	20 X1	21 V ⁽²⁶⁾	22	23	24	25	26	27 XV	28	29	30	Γ					
July				1	2	3	4 XV:	5 (28)	6	7	8	9	10	11	12 (29)	13	14	15	16	17	18 XV3	19 130	20	21	22	23	24	25	26	27	28 XVII	29 II	30	31			
August							1	2	3	4	5	6	7	8	9	10	11	12	13	14	15 XIX	(16 34	17	18	19	20	21	22	23	24	25	26	27 XX	28	29	30	31
September			1	2	3	4	5	6 (37)	7	8	9	10 XXI	11	12	13	14	15	16	17	18	19	20	21	22 XXI	23 I	24	25	26	27	28	29 XX	30 111	Γ				
October					1	2 XX1	3 11	4	5	6	7	8	9	10	11	12	13	14	15 XXIV	, 16	17	18	19	20	21	2 2 xxv	23	24	25	26	27	28	29	30	31		
November	1	2	3	4	5	6	7	8 (46)	9	10	11	12 xxv3	13	14	15	5 1 C	5 17	18	19	20	2	1 22	23 XXV	24	25	5 26	27	28	2 S (49	3 (xxy)				Res of			
December			1	2 X	3 XVI	4	5	6	7	8	9	10 XXIX	(¹¹	12	13	14	15	16	17 xxx	18	19	20	21	22	23	24	25	26	27	28	29	30	31				
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F				IV	all	ntei	nar	ice					XX	IL	vr c	cyc	e													m	gri	inż.	And	trze	j Ge	ołął	

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





FIG. 5. Diagram of MARIA RR core.



HANDLING OF IRRADIATED TARGET IN MARIA REACTOR BUILDING

- 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.
- 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.



FIG. 6. Handling of irradiated target in MARIA reactor building.









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Świerk, 03 June 2010

CERTIFICATE OF RADIOACTIVE SOURCE

no. 15/10

- 8 uranium targets / irradiation container: 29/10 and 30/10 1. Preparation:
- 2. Irradiation Rig: channel i-6
- 3. Batch: 2010.13/311205

Position holder	Target number	Position holder	Target number
	NRGA3911		NRGA3915
	NRGA3912	LIDDED	NRGA3916
LOWER	NRGA3913		NRGA3917
	NRGA3914		NRGA3918

- Mo-99 4. Radionuclide: gla-Mo · Ju 7773 Ci on EOI 5. Mo-99 activity/batch: < 410 W 6. Residual power:
- MARIANNE no. 01 7. Container type:

9. Time of irradiation:

Start of irradiation: Stop of irradiation: 27 May 2010 - at 18:20 02 June 2010 - at 14:45

11. Total

140h 25'

Laboratory Dosimetry DOZYMETRYSTA KOORDYNATOR PROGRAMÓW nisz Jaroszewicz mgr inż







CONCLUSION

- The realization of the molybdenum program confirmed the correctness
 - of the irradiation technology,
 - handling operations in the reactor pools and in hot cell,
 - loading operation into the transport container MARIANNE.
 - Experience acquired made it possible to implement additional technical and organizational solutions.
 - The achieved very good results of production is an important step in increasing of commercial products and services of MARIA research reactor.



