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Production of fission product ^{99}Mo using high-enriched uranium plates in Polish nuclear research reactor MARIA: Technology and neutronic analysis

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Abstract. The main objective of ^{235}U irradiation is to obtain the ^{99m}Tc isotope, which is widely used in the domain of medical diagnostics. The decisive factor determining its availability, despite its short lifetime, is a reaction of radioactive decay of ^{99}Mo into ^{99m}Tc. One of the possible sources of molybdenum can be achieved in course of the ^{235}U fission reaction. The paper presents activities and the calculation results obtained upon the feasibility study on irradiation of ^{235}U targets for production of ^{99}Mo in the MARIA research reactor. Neutronic calculations and analyses were performed to estimate the fission products activity for uranium plates irradiated in the reactor. Results of dummy targets irradiation as well as irradiation uranium plates have been presented. The new technology obtaining ^{99}Mo is based on irradiation of high-enriched uranium plates in standard reactor fuel channel and calculation of the current fission power generation. Measurements of temperatures and the coolant flow in the molybdenum installation carried out in reactor SAREMA system give online information about the current fission power generated in uranium targets. The corrective factors were taken into account as the heat generation from gamma radiation from neighbouring fuel elements as well as heat exchange between channels and the reactor pool. The factors were determined by calibration measurements conducted with aluminium mock-up of uranium plates. Calculations of fuel channel by means of REBUS code with fine mesh structure and libraries calculated by means of WIMS-ANL code were performed.

Key words: fission products • ^{99}Mo production • neutronic calculations • research reactor

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Introduction

Technetium-99m (^{99m}Tc) is the most frequently used radioisotope in nuclear medicine diagnostics. The half-decay period for this isotope is around 6 h. ^{99m}Tc is generated from the parent isotope of molybdenum-99 (^{99}Mo), which is generally produced in multistage irradiation processing of uranium target containing uranium-235 (^{235}U) in nuclear reactors. Annually, around the world approximately 25 million diagnostic medical procedures are performed by using ^{99m}Tc radioisotope, which constitute about 80% of all the procedures of nuclear medicine. ^{99m}Tc is generated from β-decay of parent isotope ^{99}Mo exclusively by irradiation of uranium targets in research nuclear reactors.

Currently four companies are sharing 95% of the world production of ^{99}Mo: MDS Nordion (Canada), Tyco Healthcare Mallinckrodt (Netherlands), IRE (Belgium) and NRP Radioisotope (Pty) Ltd. (South Africa). Technologies for obtaining ^{99}Mo are based on irradiation of high-enriched uranium (HEU) plates. Only 5% molybdenum comes out from irradiation of low-enriched uranium (LEU) plates. Countries implemented the production of ^{99}Mo based on LEU fuel are Australia (ANSTO) and Argentina (CNEA). In the

alternative reactor technology for obtaining ^{99}Mo, minimal amount of this isotope is originated from irradiation of ^{98}Mo plates in reaction ^{98}Mo(n,γ)^{99}Mo. This technology is used in China, India, Brazil and Kazakhstan. The technology aimed to fabricate molybdenum plates of enhanced density and higher enrichment of ^{98}Mo isotope is mainly developed in Japan (JAERI).

From the diagnostic point of view, the basic advantages of ^{99m}Tc are: short lifetime ($T_{1/2}$ = 6 h) and the emission of low energetic gamma radiation (140.5 keV). Short lifetime is, simultaneously, a drawback in relation to transport and storage of this radioisotope. In case of ^{99m}Tc, it can be dispatched and stored as a technetium generator containing ^{99}Mo mother's isotope. The ^{99}Mo has longer half-life period ($T_{1/2}$ = 66 h) and ^{99m}Tc can be produced from it. The isotope ^{99m}Tc can be eluted from generator using saline right before the examination. Usage of ^{99m}Tc for a single examination does not exceed ca. 50 μCi (1.85 MBq) for 1 kg of patient's mass.

Production of ^{99}Mo by means of carrier method (i.e. ^{98}Mo target irradiation) was carried out in EWA and MARIA reactors within 1970s and 1980s. During 1980s, the fuel element of WWR-SM type in the EWA reactor was irradiated and transported to Rossendorf to release the isotope ^{99}Mo. The first trial over the irradiation terms at MARIA reactor of uranium plates to produce ^{99}Mo was conducted at the beginning of 1990s [1].

At the initial point, the choice of technology for obtaining ^{99}Mo and selection of target material are the technical difficulties faced. To resolve this, two technologies were followed: (1) procedure based on reaction (n,f), in this the source for ^{99}Mo is the fission products of ^{235}U or (2) ^{98}Mo is produced by neutron capture reaction (n,γ) during irradiation of targets of natural or enriched one by the isotope ^{98}Mo, mainly in the form of targets containing MoO_3.

Assuming that it is aimed to achieve the possible highest specific activity of ^{99}Mo is the preferable technology to gain ^{99}Mo from the fission products of ^{235}U after irradiation of uranium targets. The main producers of highly enriched uranium targets belongs to the French company CERCA offers the uranium targets from ^{235}U dispersed in aluminium: UAl_xAl as well as oxide targets containing uranium in the form of UO_2. Keeping this in mind, technology for the irradiation of targets in MARIA research reactor has been developed.

Production of ^{99}Mo from uranium fissions

The isotope ^{99}Mo forms within the chain of β-decays of consecutive fission fragments of short life originating from ^{99}Rb. The cumulated productivity of ^{99}Mo by one fission is γ = 0.0611. All fission products leading to the formation of short-lived ^{99}Mo, the most long-lived isotope ^{99m}Nb has a half-life period 2.6 min. By considering the longer half-life period for ^{99}Mo than radionuclides leads to the assumption that ^{99}Mo rises immediately as a product of ^{235}U fissions. Under such assumption the rate of building up of ^{99}Mo activity in determined portion of fuel is proportional to the fission power released. If we assume that the energy released during one fission is $E_r = 3.244 \times 10^{-11}$ J and the decay constant of ^{99}Mo is $\lambda = 2.9 \times 10^{-6}$ s^{-1}, then the factor of proportionality between the build up of ^{99}Mo activity rate and fission power is:

$$\alpha\lambda/E_f = 5.47 \times 10^3 \text{ Bq/Ws} \tag{1}$$

In the course of uranium plate irradiation, the ^{99}Mo activity changes over the exposition time in accordance with the equation:

$$\frac{dA(t)}{dt} = \alpha P_f(t) - \lambda A(t) \tag{2}$$

where: $A(t)$ – activity of ^{99}Mo in the course of irradiation; $P_f(t)$ – fission power generation by uranium plate.

Integrating Eq. (2) within the limits over the plate irradiation time $(0,t)$, we receive a relation activity of ^{99}Mo formed from the fission power generation in the plate:

$$A(t) = \alpha \int_0^t P_f(\tau) e^{-\lambda(t-\tau)} d\tau \tag{3}$$

This relatively simple relation constituted the basis for determining the quantity of pending ^{99}Mo to be generated during the irradiation of uranium plates in the reactor. It is the basis for the development of irradiation technology for uranium plates in MARIA reactor. It has been directly used as a relation between the fission power generated in uranium plates irradiated and the activity of ^{99}Mo formed. The relation (3) enables to perform a continuous control activity of ^{99}Mo produced on a condition that the fission power $P_f(t)$ released in a continuous way to the irradiated target is monitored. Such possibility is ensured by specific construction of MARIA reactor core as well as ensured by introducing specific equipment along with SAREMA measuring system [2]. The power measured by a reactor measuring system does not precisely correspond with the fission power generated in uranium plates. This measurement have been burdened by certain systematic errors of which the major fraction occurring on the power due to the impact of gamma radiation emitted by the neighbouring fuel elements and heat removal through the reactor pool. To define the impact of those effects and to obtain effective fission power by implementing correction factor have to be assessed on experimental way.

Technology of uranium target irradiation in MARIA reactor

Technical characteristic of uranium targets

The uranium targets in the form of plates with dimension of 203 × 40 × 1.45 mm with the core including fission material ^{235}U in the shape of dispersion in aluminium: UAl_x ($x \cong 3$) have been assigned for irradiation. Fission material of the core is positioned between to aluminium plates (cladding) contains high-enriched uranium in which the ^{235}U contents is over 90%. In commonly used nomenclature there are so-called HEU (high-enriched uranium) targets. Such composed sandwich is then hot and cold-rolled. After rolling on cold and verified the dimensions of the fuel core by use of fluoroscopy method, the edges of uranium plate are cut to match the dimensions and subjected

Table 1. Technical characteristic of uranium plates

Parameter	Value
External dimensions of uranium plate	203 × 40 × 1.45 mm
Dimensions of fuel core (mean)	186 × 29.5 × 0.85 mm
Uranium mass in plate	4.7 g (4.61÷4.79 g)
Tolerance of uranium-235 mass in plate	±2%
Enrichment	89÷93%
Material of fuel core	Dispersion UAl_x ($x \cong 3$) in Al
Cladding material	Aluminium alloy 1050A (BS)

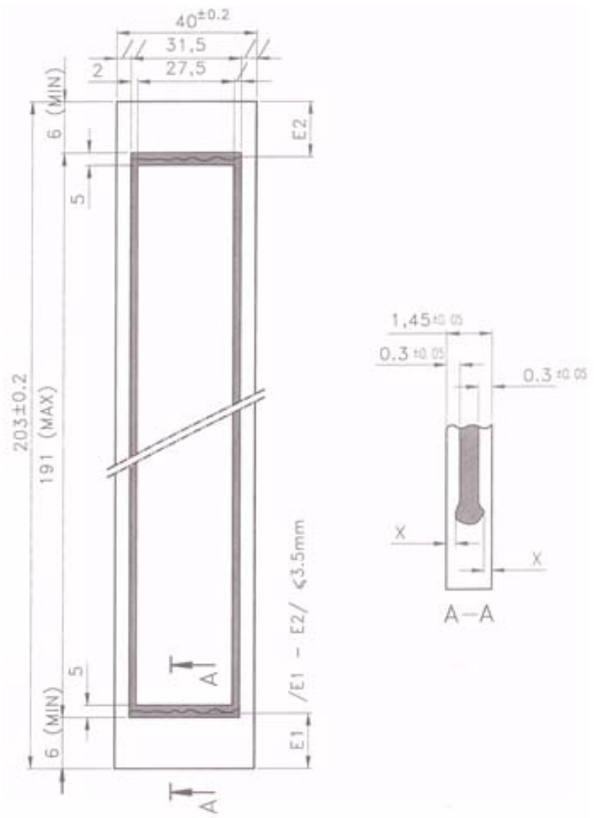

Fig. 1. Uranium target of HEU.

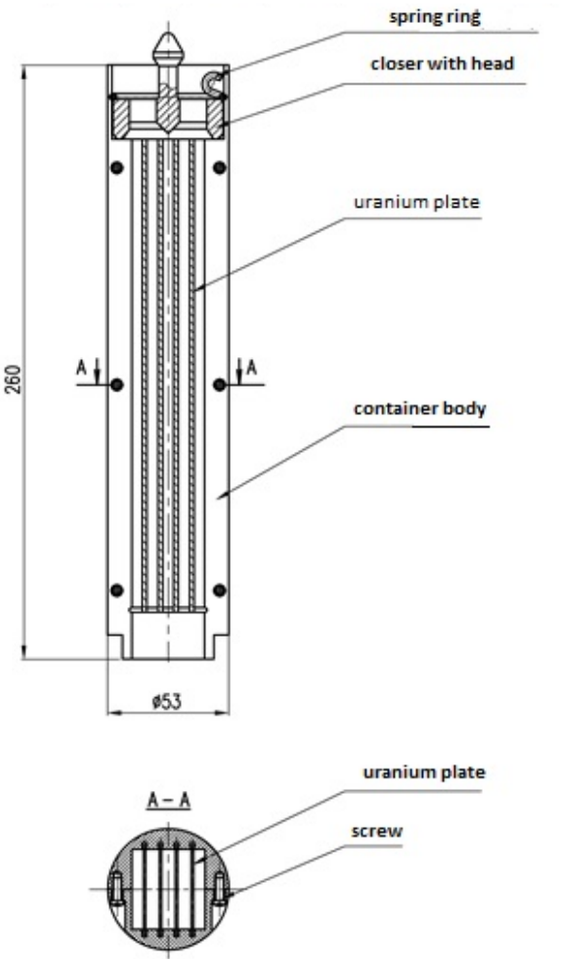

Fig. 2. Container for irradiation.

to the number of tests. One of the tests to investigate the uranium is a 'blister test'. During this investigation the plates are subjected to annealing in 425°C for 1 h. From the viewpoint of reactor technology it is great importance to check the uniformity of uranium distribution as well as the thickness uniformity of the layer of cladding. Another important parameter is the fuel cladding quality. Precise observation should, especially, be turned on cracks or pits. Possible faults are directly link with decreasing effectiveness, to be one of the barriers limiting the propagation of fission products being generated. Table 1 includes technical characteristic of uranium plates used for irradiation. The sketch of target presents in Fig. 1.

Conditions of the irradiation of uranium plates

Typical container for irradiation of uranium target includes eight plates positioned in special cans, four plates on each one (Fig. 2). The set of eight plates constituted charge of individual channel for irradiation placed in central zone of reactor core. Configuration of reactor core with the fixed position, i-6 and f-7, channels to be used for irradiation of uranium targets (Fig. 3). The length of active region of MARIA reactor core is 1000 mm; length of irradiated plates is a bit over 200 mm brings about more than 40% of the available loading volume can be used. Hence, it is reasonable to consider the procedure over the simultaneous irradiation of the three cans containing 12 uranium plates altogether (Fig. 4). Relating to the vertical distribution of the thermal neutron flux density in molybdenum channel increases the production output of ^{99}Mo by around 37%, if we use the option with three cans [3].

Technology for the irradiation of uranium plates with the goal fuel element of MARIA reactor has taken place (Fig. 4). Reactor fuel channel used to irradiate uranium plates relies on its modification to provide multifold loading and discharging of uranium sets with targets in and out of installation without the necessity

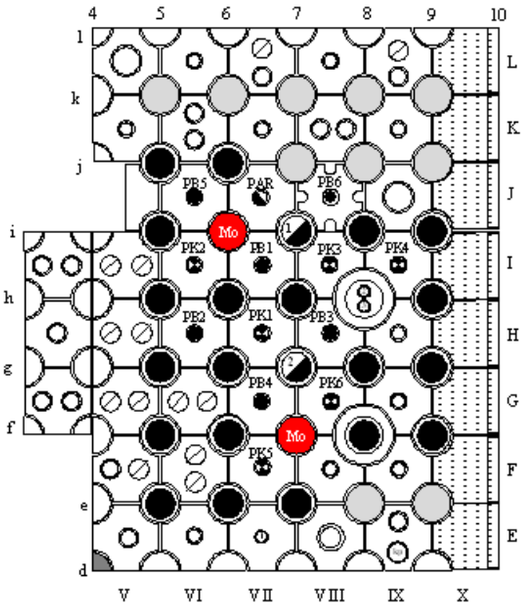

Fig. 3. Configuration of the MARIA reactor core with molybdenum channels positions i-6 and f-7.

of removing the whole molybdenum channel from the position socket in the reactor core. The construction of the internal fuel structure allows to removing the fuel channel head after the reactor shutdown and transported the irradiation container together with uranium targets beyond the hoop of reactor core. It is in line with a standard procedure adhered for the reactor fuel channel. The assumed technology of uranium target irradiation provides the possibility to use the standard reactor channel cooling system for the removal of heat from molybdenum channels. The thermal power generated in the eight uranium plates, determining typical set to be irradiated, does not exceed 200 kW and in case of the set of 12 plates it will achieve ca. 300 kW. The possibilities of the heat removal from an individual fuel channel of MARIA reactor under nominal parameters of the coolant flow are significantly greater. It should be remembered that due to, relatively, low thermal power generated in uranium plates, the thermal heat flux on the plate's surface is greater than the thermal heat flux on the surfaces of the other operated fuel elements in the MARIA reactor. Values has been compared with data for fuel elements MR type (36% enrichment in isotope ^{235}U) or for the new low-enriched fuel MC type elements (enrichment 19.75%).

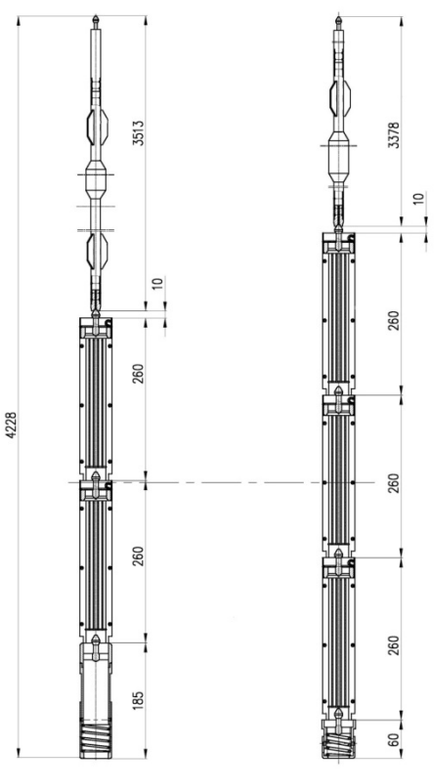

Fig. 4. Two options over molybdenum channel – with the two sets (2 × 4 targets) and three sets (3 × 4 targets of HEU).

Due to the high thermal flux on plate's surface and the possibility of changing the position of reactor core of the installation for irradiation, it is necessary to preserve the nominal coolant flow. Differences in maintaining the rate of nominal coolant flow in the channel for the irradiation due to inaccurate measurement of temperature has impact in measuring the power generated in the channel, but at the same time it allowed to perform more precise calibration of the measuring systems to calculate the generated power by the set of uranium plates.

Pressure drops on the sets with uranium plates are lower than in case of the MR and MC fuel elements used in MARIA reactor. Admittedly the standard fuel channels are equipped with outlet valves to control the coolant flow rate through the fuel channel, but in case of uranium plate irradiation the throttling of flow by means of the outlet valve seems to be deceptive. Therefore an additional gland seal to increase the hydraulic resistances has been installed at the channel outlet section, which eliminates the potential threat compared with MR type fuel elements [4].

Cooling and transportation of irradiated uranium targets

Because of high heat flux and possibility of changing the position in reactor core of the installation to irradiate uranium plates, it is reasonable to prevent the rated coolant flow rate in the installation. It is true that introducing the nominal coolant flow rate in the channel during irradiation worsen the precision of temperature difference measurement, however it enables to provide more precise calibration to measure the power of plate's compound.

Pressure drop of water on the irradiated cans with uranium plates are substantially lower in MARIA reactor operated with fuel elements MR and MC type. The standard reactor fuel channels are equipped with outlet valves to control flow rate through the channel. In the case of channels used for irradiation of uranium plates the tottering of flow by means of outlet valve may be uncertain. The problem can be solved by upgrading the construction to eliminate potential threat of the channel aimed to irradiation by means of applying an additional component with increasing hydraulic resistances in the outlet part.

Measurement of temperatures and flow rate of coolant in molybdenum installation are continued by means

of standard reactor measuring system SEREMA, however they are not providing direct information on updated fission power generated in uranium plates. To do the procedure of bretton power measuring system, it is required to perform the designed installation an irradiating of the set of aluminium mock-ups of uranium plates.

Preliminary cooling of uranium plates when terminating the irradiation is being done in installation for irradiation within the framework of procedure over removal of the shutdown residual heat to be generated in fuel of MARIA reactor. This cooling is provided by operation of reactor fuel channel cooling system. Unloading of cans with uranium plates can be done after disengaging reactor cooling system and closing the valves in molybdenum installation. The unsealing of molybdenum installation and evacuation of cans with uranium plates will be carried out after around 10 hours from terminating the irradiation. The thermal power to be generated in the set of eight uranium plates will be after such period of cooling ca. 500 W.

A consecutive stage of technology is to transport of cans with uranium plates from installation in the reactor core to the reactor-dismantling cell. It is connected with change of cooling conditions of cans with uranium plates. As the reloading operations at the dismantling cell will be transition from cooling in water medium to convection cooling in the air.

Cooling of plates by means of natural convection in the air is decisively less efficient than convective cooling in water. Therefore it is necessary to determine the thermal power generated in the cans with plates and to compare with maximum value of power to be admissible for cooling conditions in air. The receiver of uranium plates are the two thermal limits for the set of the eight uranium plates: power 548 W during that one can start transporting from dismantling cell to loading into special transport container with power 450 W is a maximum allowable value to transport the container on public roads. This limit is related to the activity of plate sets strongly correlated with thermal power. Before commencing transport operation over the cans with plates in air, the calorimetric measurement of heat generated in individual can with plates will be performed. The measurement will be done in the reactor storage pool.

Due to uncertainty associated with the calculations of heat exchange under natural convection in air, it is necessary to conduct temperature measurement in uranium plates at the dismantling cell and on that basis to determine power limit, under which one can safely carry out the reloading operations in the dismantling cell.

Overall activity of fission products in uranium plates pending transport operations at the dismantling cell is order of 100 kCi [5]. In this connection the shielding capacity of dismantling cell has to be verified in both regarding the calculation and experimental procedure and the data obtained. Above all it is importance to retain the nominal level of water storage pool (+7.05 m), that enables immersion in water of the front wall of dismantling cell. The stages of reloading irradiated targets procedure are presented in Fig. 5.

Determining the activity of ^{99}Mo in irradiated uranium targets

The activity of ^{99}Mo in irradiated uranium targets is generally determined by using the data collected by reactor measuring system SAREMA. This system is recording data concerning the coolant temperature at the fuel channel inlet and outlet ($T_{in,k}$, $T_{out,k}$ respectively), water temperature in the reactor pool ($T_{in,b}$) as well as flow rate in the channel (G_k). Using the quantities measured the total reactor power (P) to be sum of thermal powers of fuel channels and reactor pool is calculated. Recording of the above mentioned data is taken with a frequency

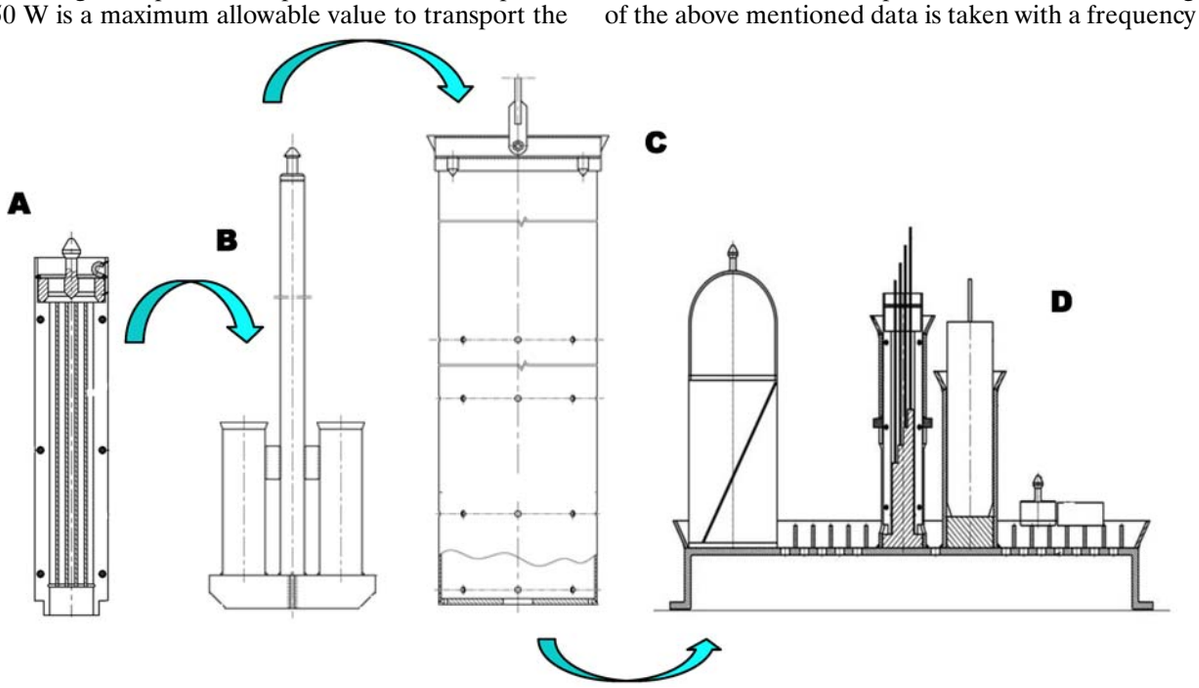

Fig. 5. Procedure of reloading operation with irradiated uranium targets in MARIA reactor. (A) – irradiation container in molybdenum channel; (B) – two-positional transport container; (C) – inside pool transport facility; (D) – reloading stand inside the disassembling (hot) cell.

of 1/s. Hence, one can assume, that the system deliver continuous information over the reactor power and the power generated by individual fuel elements. These measurements however, do not supply direct information on current fission power generated in fuel channel, because they are taking into account additionally the heat generation due to gamma radiation from neighbouring fuel elements and the heat exchange between fuel channels and reactor pool in power balance.

The system SAREMA has been adapted to servicing the process of uranium targets irradiation in MARIA reactor as an effective tool aiding the operator in evaluation of thermal hydraulic parameters of reactor molybdenum channels. The feature of the technological channels in process uranium targets irradiation requires introducing certain changes into the system, which is highlighted in details, in the internal report [6].

To estimate the ^{99}Mo activity in molybdenum channel it is necessary to get information on real fission power to be generated in uranium plates. It requires standard power measurement recorded by SAREMA system a portion of external gamma radiation as well as an effect of heat exchange between channel and the reactor pool.

$$P_f = P_S + P_{cor} \tag{4}$$

where: P_f – real fission power to be generated in molybdenum channel, P_S – thermal power measured by the SAREMA system, P_{cor} – correction introduced.

Similarly as for ordinary fuel channel also for the molybdenum channel the cooling temperatures at the channel inlet and outlet ($T_{in,k}$, $T_{out,k}$), reactor pool water temperature ($T_{in,b}$) and coolant flow rate at the channel (G_k) are recorded in the SAREMA system. Also in the SAREMA system is counted the sum of fuel channel thermal power (ΣP_k). Taking into account the thermal-hydraulic parameters in the equation of reactor heat balance, the fission power generated in the molybdenum channel during irradiation of uranium plates to be measured by the SAREMA system in the given moment of time is calculated by Eq. (5):

$$P_S = \lfloor (\rho c_p T)_{out,k} - (\rho c_p T)_{in,k} \rfloor G_k = P_f - A_{kb}(T_{in,k} - T_{in,b}) + \gamma_m P_k \tag{5}$$

A_{kb} – heat transfer coefficient from the fuel channels to the reactor pool, γ_m – coefficient determining the fraction of thermal power due to gamma irradiation in uranium targets.

The implemented correction factors A_{kb} and γ_m are experimentally defined by irradiation of the sets of aluminium mock-ups of uranium plates [7]. The irradiation of a plate that does not contain fission material in molybdenum channel allows to assume that the fission power to be generated in the channel is equal $P_f = 0$. By applying these assumptions into Eq. (5), it is possible to define the introduced correction which has the form:

$$P_{cor} = A_{kb}(T_{in.k} - T_{in,b}) - \gamma_m \Sigma P_k \tag{6}$$

The correction element uses the global parameters recorded by the SAREMA system. They are above mentioned: water temperature at the inlet of molybdenum channel circuit, water temperature at the inlet to reactor pool ($T_{in,k}$ and $T_{in,b}$) and the total power from the fuel channels (ΣP_k). The results ensured to calculate the correction factor values in the equation binding activity of the ^{99}Mo to be raised with the fission power generated in uranium plates. For the fixed thermal powers, sum of the fuel channels (ΣP_k), the factors A_{kb} and γ_m it is possible to assess experimentally by matching the straight line into the measured values ($T_{in,k} - T_{in,b}$) as well as P_S. The value of factors for molybdenum channel with two irradiation containers, i.e. with the eight uranium plates are respectively:

$A_{kb} = 4.82 \times 10^{-3}$ [MW/deg], $\gamma_m = 1.29 \times 10^{-3}$.

Activity variation of ^{99}Mo during irradiation is calculated by means of formula derived from rearranged (converted) formula (3):

$$A(t + \Delta t) = A(t) \cdot \exp(-\lambda \Delta t) + \alpha \cdot \Delta t \cdot \frac{P_f(t) + P_f(t + \Delta t)}{2} \tag{7}$$

where: $A(t)$ – activity ^{99}Mo for given moment of time, λ – ^{99}Mo decay constant, α – factor of proportionality between speed acceleration activity of ^{99}Mo and the fission power for an individual fission, P_f – fission power.

Figure 6 shows the screen of SAREMA system along with visualization of thermal-hydraulic fuel channels of MARIA reactor as well as visualization of ^{99}Mo activity level created as a result of irradiation process over uranium targets in molybdenum channels: i-6 and f-7.

The activity of ^{99}Mo in uranium plate is a function of the fission power generated in the fuel $P_f(t)$, irradiation time t_a and its cooling time t_c can be computed by means of code ORIGEN [8] or it can be calculated from modified relation (3) in the form:

$$A(t_a, t_c) = \alpha \int_0^{t_a} P_f(\tau) e^{-\lambda(t_a - \tau)} d\tau \cdot e^{-\lambda t_c} \tag{8}$$

where: $A(t_a,t_c)$ – an effective activity of ^{99}Mo after irradiation and cooling, $P_f(\tau)$ – fission power generated by the uranium plate, t_a – irradiation time of uranium plates, t_c – cooling time of uranium plates, α – factor of proportionality equal 5.47×10^3 Bq/Ws.

The reference value of ^{99}Mo activity equal 6.67 kCi (2.46×10^{14} Bq) has been taken for the initial analyses. The calculations exhibit that to achieve such activity, one has to irradiate a set of eight uranium plates with a mean fission power 188 kW for 150 h. A comparison of the results gained by means of ORIGEN code with data based on assessment activity by performing the measurement of fission power in the molybdenum channel exhibits a large convergence. Hence, for instance the activity of ^{99}Mo determined from the calculations by means of ORIGEN code to be referred to the real conditions of irradiation, i.e. 144 h and the fission power 175 kW is lower by around 3.1% in comparison with activity ^{99}Mo calculated according to Eq. (8). Validity of employing in irradiation analyses in MARIA reactor the method for determining the activity of ^{99}Mo as a direct function of the fission power have additionally confirmed the activity measurement of ^{99}Mo performed in the processing laboratory in the Netherlands. The measurement of activity gained from the fission product of ^{99}Mo being conducted after the cooling time in reactor and transport time was equal in total ca. 44 h and after the conversion time of the irradiated plates in the

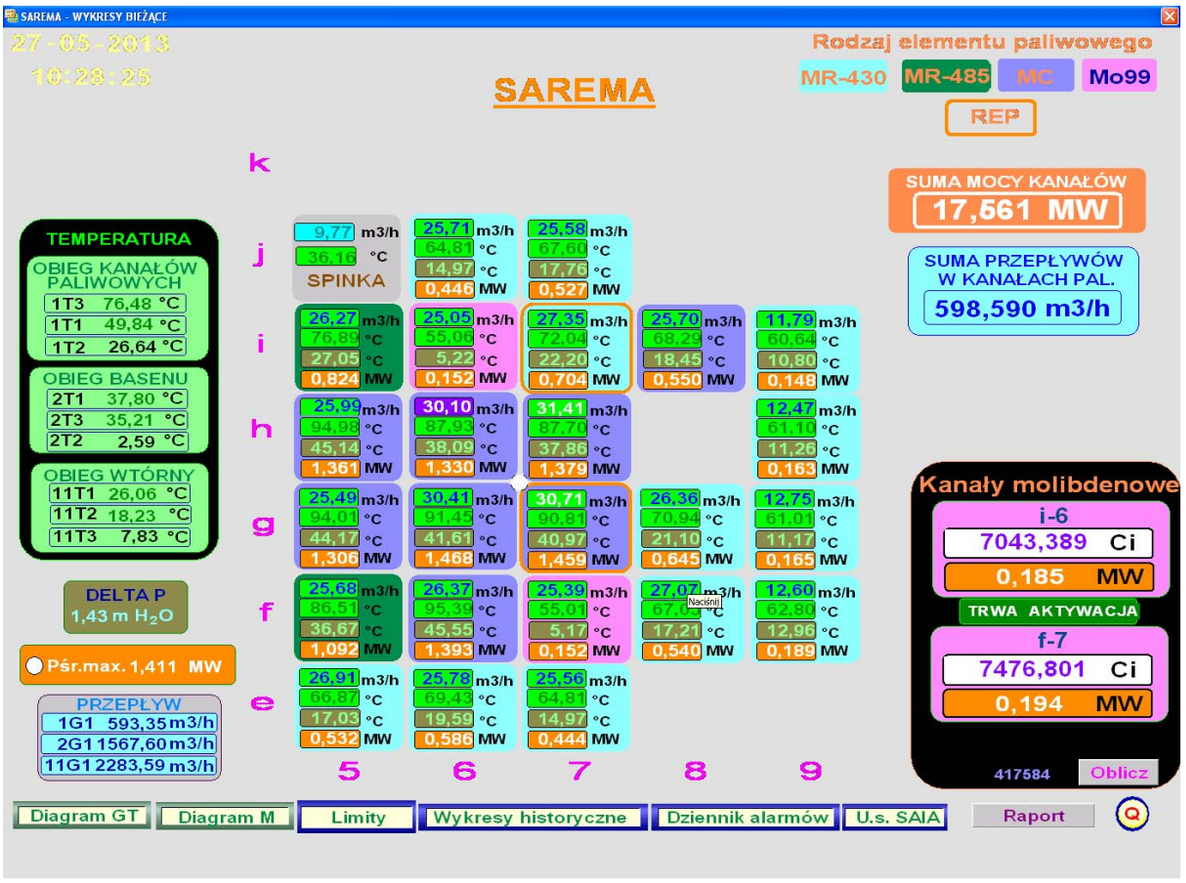

Fig. 6. Screen over SAREMA system – visualization of thermal-hydraulic parameters of MARIA reactor.

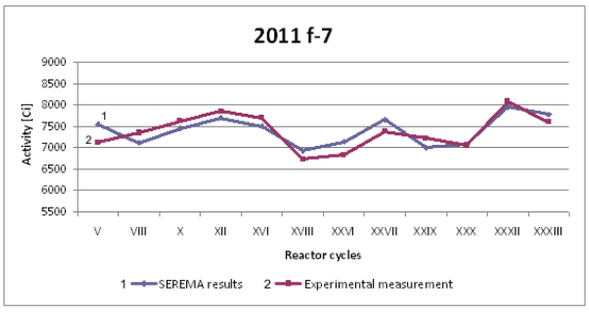

Fig. 7. Activity of ^{99}Mo computed according to procedure binding activity with fission power and the experimental data from the measurement at the Laboratory in Petten (data are referred to the molybdenum channel in position f-7).

laboratory gives the results differing between one and the other by around 3.5%. If we relate these results to the time over terminating the irradiation, i.e. EOI (end of irradiation) and taken into account the capacity of converting process of irradiated uranium plates at the Laboratory in Petten. It is worth to highlight that the convergence of the results is satisfactory. Exemplified data are presented in Fig. 7.

Neutron calculations

The principal goal over the neutron calculations of the MARIA reactor core was the choice of location of the channels for irradiation of uranium plates. The calculations were aimed to determine the reactivity excess and the thermal power generated in uranium plates for various positions of the molybdenum installation in the reactor core. An assumption has been taken into mind that the calculations are conducted for the two parallel operating installations for irradiation of uranium plates.

For the neutron calculation of the reactor core with installation for irradiation of uranium plates has been taken into consideration as the reference reactor core configuration as shown in Fig. 8. The physical calculations have been performed by means of the standard calculation codes being applied in analyses of neutron parameters of the reactor core. This can be used as the reactor codes as follows:

- code WIMS-ANL employed for the cell calculations and was prepared by the library for active cross-sections of installation cells to irradiating the uranium cells [9];
- diffusion code REBUS with a new library of active cross-sections [10];
- code Monte Carlo for calculations and distribution of neutron flux in the core [11].

The main calculations aimed to define potential location of molybdenum installations in the reactor core were performed by means of the diffusion code REBUS [8]. Conclusions from these calculations are as follows:

- Based on the calculation the selected positioning must foremost provide to generate appropriately high thermal power in uranium plates. The power

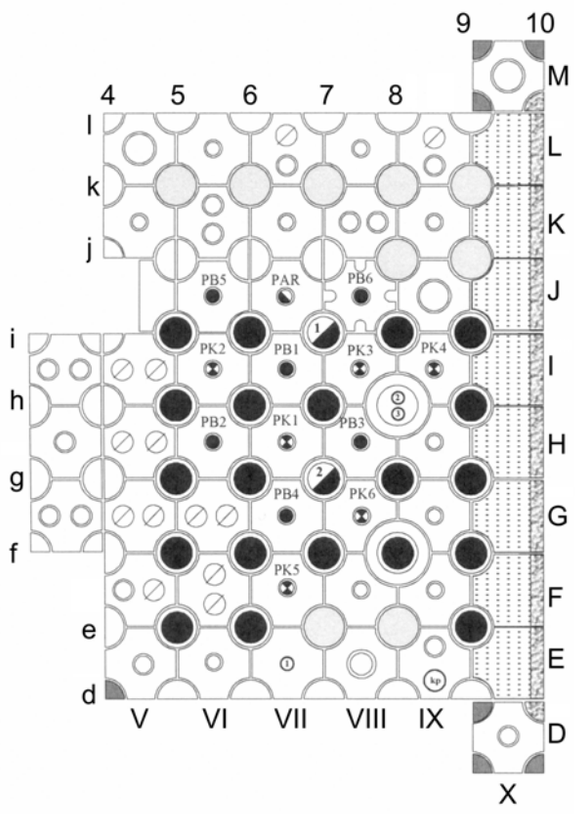

Fig. 8. Reference MARIA reactor core used for neutron calculation.

level of 0.19 MW generated in plates is attainable only when the installation is positioned in the central area of the reactor core. An additional requirement is to locate the molybdenum installation in the vicinity of low irradiated fuel element of MR or MC type of thermal power 1.8 MW which brings about that positioning of installation for irradiation of uranium plates in the central zone of the core substantially lowers the reactivity excess of the core. To compensate this effect, it is necessary enlarge the core and/or operating it with larger amount of ^{235}U.

– Calculations unveiled a strong dependence of thermal power generated in uranium plates from configuration of fuel elements in the reactor core as well as of power and fuel burn up level in the vicinity of molybdenum installation.

Configuration of MARIA reactor core with two molybdenum installations in core-positions h-7 and f-7 presents Fig. 9.

To provide an appropriate reactivity excess, in comparison with reference core the fuel element was placed in position e-7 and the central fuel channels (positions h-6, g-6 and f-6) were made up with fresh fuel elements. The core contained over 120 g ^{235}U more (in standard reactor nuclear fuel type MR or MC) in comparison with a reference core. This made to replenish the reactivity excess is different for each configuration but bearing in mind the abovementioned example, one can evaluate that 'the reactivity costs' for placing two molybdenum installations in MARIA reactor are equivalent to an additional fuel element MR or MC of medium burn-up.

Fig. 9 also shows graphic power density distribution in the uranium channels of MARIA reactor core. Numerical values of power are collected in Table 2.

Location of the two molybdenum installations in the MARIA reactor core does not significantly affect the magnitudes of reactor kinetics parameter. An effective delayed neutrons share is $\beta_{eff} = 6.9 \times 10^{-3}$ and contained within the limits of variability of that coefficient [2]. It is only to be observed insignificant growth of effective lifetime of neutron generation up to the

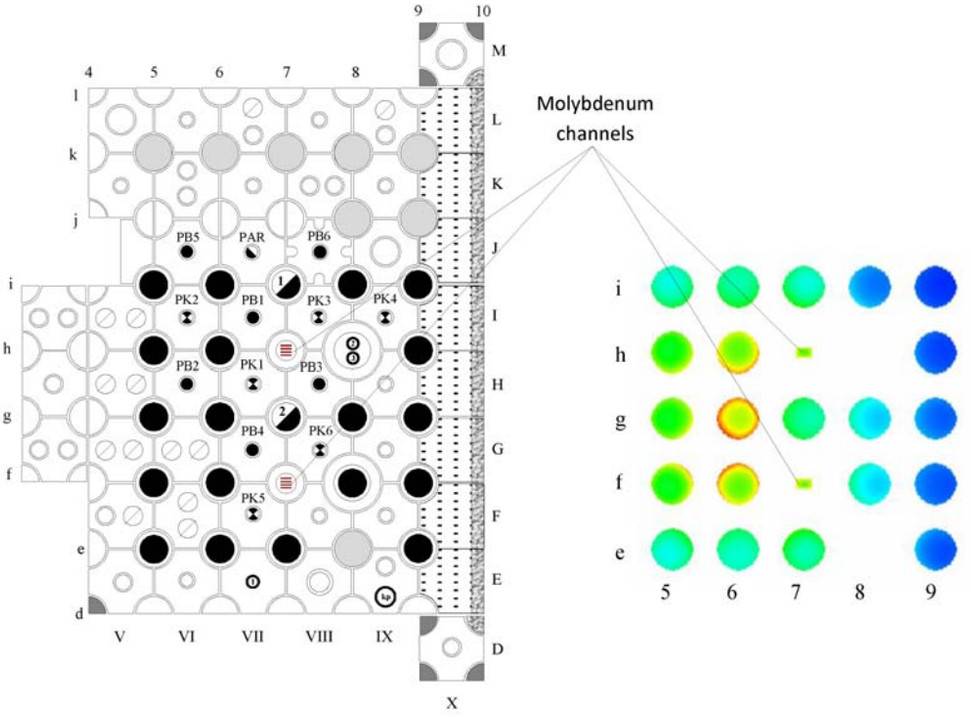

Fig. 9. MARIA reactor core configuration with two molybdenum installations.

Table 2. Thermal power individual fuel elements [MW] in configuration from Fig. 9

	5	6	7	8	9
i	0.89	1.00	0.94	0.39	0.17
h	1.44	1.75	**0.19**	no fuel	0.23
g	1.44	1.66	0.97	0.65	0.32
f	1.49	1.80	**0.19**	0.69	0.28
e	0.89	0.90	1.03	no fuel	0.30

value $\Lambda_{eff} = 155$ μs (in MARIA Reactor Safety Report, ERB [2] is taken the value $\Lambda_{eff} = 144 \pm 4$ μs), which is resulted from changes of fuel lattice after installing the molybdenum channels.

The neutron calculations are conducted by means of MCNP code to determine unevenness degree of power density distribution in uranium plates. There are four main sources for this unevenness: axial density of thermal neutron flux, horizontal distribution in beryllium matrix, microscopic distribution of neutron flux in the can with plates and irregularity of fuel distribution in uranium plates.

Due to insignificant burn-up of uranium plates the axial distribution of power density does not substantially differ from an axial density of thermal neutron flux and with a good approximation it may be deserted by a relation:

$$q_v(z) = q_v^{\max}\cos(\pi/\hat{H}) \tag{9}$$

where $z = 0$ corresponds to a half-length of the core, $\hat{H} = 1.15$ m is to called extrapolated length of the core. This relation (9) will be further used in the calculations.

The irregularity of horizontal distribution of flux density in beryllium matrix depends on molybdenum installation and its orientation regarding the use and one should treat them as irregularity of a random nature. In positions h-7 and f-7 there have not been noticed irregularities surpassing +10%.

Microscopic distributions of power density in uranium plates exhibit the presence of systematic effect of neutron self-screening. The outer plates from the batch of four plates in the can unveil power densities by +11% larger than the power densities in the inner plates. The irregularities of fuel distribution in plates are of random nature and do not surpass ±20%. This conforming that the power density irregularities are lower than ±20%.

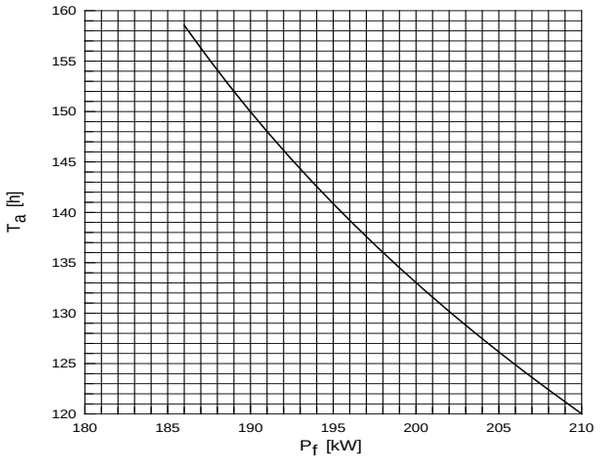

Fig. 10. Relation between irradiation time (T_a) of uranium targets and generated fission power (P_f).

The procedure of reactor start-up with molybdenum irradiation channels, loaded with uranium targets, assumes gradually increasing of reactor power up to reaching generated fission power in the channel on range 185÷210 kW. Within these ranges the power of reactor fuel elements do not exceed nominal power on range 1.3÷1.8 MW. Relation between irradiation time of uranium targets and generated by uranium targets fission power shown in Fig. 10. Relation presented in Fig. 10 is helpful for monitoring ^{99}Mo production procedure.

Discussion and summary

Technology of obtaining the ^{99}Mo from the fission products is one of the most effective methods for gaining this isotope. It enables to gain the isotope of high specific activity, which is a very important property from the viewpoint over the production of molybdenum-technetium generators commonly used in oncological diagnostics.

Introduced technology for irradiation of uranium targets in the reactor is relatively simple way of obtaining ^{99}Mo. It is very effective utilization of the MARIA reactor structured possibilities and comes up to the fundamental relationships between the fission power generated in the targets and the activity of the created ^{99}Mo, one of the ^{235}U fission products.

In majority of technical solutions in other reactors, the irradiation technology of the uranium targets is based on irradiation in autonomous installation with an individual cooling system. These solutions are approximate to irradiation of target material in the typical isotope channels. The solution implemented in MARIA reactor employs a specific construction of fuel channels with the cooling system, mechanical hardware and measuring system, linked to the measuring system of thermal-hydraulic parameters of the reactor fuel elements.

The most substantial parameter, from the view of irradiation process, is activity of the arising ^{99}Mo. Generally the activity of fission products determines by the ORIGEN code. Practically the activity of ^{99}Mo is being verified only post-factum by measuring the activity separated from the fission product ^{99}Mo after termination of radiochemical processing of irradiation targets. Technology introduced in MARIA reactor assumes that activity of ^{99}Mo is directly estimated from the fission power generated in uranium targets. Such a possibility enables system SAREMA connected to the serving of irradiation process over irradiation of uranium targets. The fission products emitted in uranium targets is being assessed by means of standard power level assessed by this system. The precise assessment of the fission power additionally requires to take into account outer gamma radiation as will be the effect of heat transfer between the molybdenum channel and the reactor pool in total heat balance of the channel. The correction elements considering the above mentioned effects makes it possible to define accurately the fission power generated by

plates and in consequence based on Eq. (3) to determine the ^{99}Mo activity.

Applying the standard technological measurements during the irradiation cycle to control process by the regulation of generated power in molybdenum channel. Finally it has effect for control the amount of arriving ^{99}Mo defined by its activity. Irradiation of the set eight uranium targets of HEU inducing an average fission power of 165–190 kW over hours allows gaining at the end of irradiation activity on the level of 6800–7200 Ci (250–265 TBq). The highlighted technology to obtaining ^{99}Mo in MARIA reactor inclusive of the satisfied procedure to determining and checking activity of that isotope in the process of irradiation of uranium targets creates the unique manufacturing possibilities. The data presented in Fig. 7 show high compatibility of data to be determined in the course of irradiation process of the uranium targets in MARIA reactor with data of measurements performed at the Laboratory in Petten (Netherlands). The activity of ^{99}Mo determined after radiochemical processing irradiated uranium targets related to the activity over the EOI time points out the conformity with an error ±5%.

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